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ACIDIC PRECIPITATION IN ONTARIO STUDY (APIOS)

ANNUAL PROGRAM REPORT 1989/1990

APRIL 1991

Ontario
Environment
Environnement

ISSN 0824-880X
ISSN 0835-4456

ACIDIC PRECIPITATION IN ONTARIO STUDY (APIOS)

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Report prepared by:
The Acidic Precipitation Office
Ontario Ministry of the Environment

APRIL 1991

PRINTED ON RECYCLED PAPER
IMPRIMÉ SUR DU PAPIER RECYCLÉ

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PIBS 1534

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Introduction

In the mid-seventies, results from the Ministry of the Environment Sudbury Environmental Study and Lakeshore Capacity Study demonstrated the importance of long range transport of acid rain precursors and the negative impacts of acidic deposition on the environment. In 1979, the Acidic Precipitation in Ontario Study (APIOS) was launched to further study and document the effects of acid deposition, and to implement an effective abatement strategy. APIOS operates on a 5-year planning cycle and the second five year plan (1986-1990) has been completed. A third five year plan (1991-1996) has been developed and includes global warming and air toxics programs.

The APIOS program has four major components; scientific research, abatement, communications, and litigation. The APIOS Office is responsible for the co-ordination of these components within Ontario, at the federal level and internationally. The Office also provides administrative support to the APIOS program and has been involved in coordinating the review of a green paper on global warming and providing input into the Environmental Assessment review of Ontario Hydro's Demand/Supply Plan.

The scientific research component is divided into six tasks and its programs are developed and implemented through six interbranch or interministerial working groups.

Task 1: Atmospheric Processes Studies

Task 2: Aquatic Effects Studies

Task 3: Terrestrial Effects Studies

Task 4: Biogeochemistry Studies

Task 5: Environmental Management and Economics Studies

Task 6: Laboratory Support Services and Methodology Studies

The credibility of the scientific research is assured by a documented and operational quality assurance program.

Ontario's research and emission control efforts are coordinated with other parts of Canada and the United States since the solution to the long range transport of air pollutants (LRTAP) requires action by all jurisdictions. The APIOS program does not have a health effects or materials damage component as these are addressed by small federal specialist groups on a Canada-wide basis. The APIOS Office endeavours to keep abreast of developments in these two areas and ensures that APIOS technical support is provided when needed (e.g. deposition data).

This report will describe the programs and provide accomplishments and major findings in the six scientific work groups and provide background on APIOS abatement, communications and litigation initiatives for the period April, 1989 to March, 1990.

Appendix I provides a summary of international LRTAP projects with MOE involvement. Appendix II provides a bibliography of APIOS-related publications and technical reports.

Atmospheric Processes Studies (Task 1)

Contact: D. Yap

There are five subtasks in the Atmospheric Processes Studies program: Emissions Inventory, Modelling, Deposition Monitoring, Oxidants Strategy and Meteorological Studies.

A. EMISSIONS INVENTORY

The compilation of statistics on the production of sulphur dioxide (SO_2), nitrogen oxides (NO_x) and other pollutants serves several purposes:

a. trends in emissions of acid producing gases are determined and matched with changes in deposition patterns;

b. detailed information on SO_2 and NO_x and volatile organic compounds (VOC) emissions by geographic location is provided and required by all of the atmospheric models;

c. knowledge of the location and magnitude of emission sources is obtained which is essential in planning cutbacks of acid gas emissions.

The Ontario Acid Rain Emission Inventory has achieved its goals to include other pollutants such as ammonia, alkaline dust, cadmium, arsenic, manganese, and iron during the period 1986 to 1990. In order to maintain a consistent emission methodology, the standard EPA Source Classification Code (SCC) coding system was adopted to facilitate the breakdown of VOC species and analysis of Ontario, Canada and U.S. emissions during FY 89/90. The 1985 base year, previously updated with Mobile 3C (Canadian version of U.S. EPA transportation model for vehicles) factors for the transportation sectors, was revised in cooperation with Environment Canada using the latest factors of Mobile 4C. Improved information on solvent usage was also included. Figure 1 provides the most current information on the 1988 Ontario base year emissions for SO_2, NO_x and VOCs. An industrial survey, expanded from the 1985 data base to cover more potential sources, was carried out to collect data for 1986 and 1987. The results are being finalized and integrated into the emission inventory system. Surveys for 1988 and 1989 are currently being undertaken.

A Fast Reference Emission Document (FRED) was compiled from several sources to provide timely emission information covering Ontario and North America. A regular schedule to update and revise this document with the latest emissions information available is planned.

The preliminary SO_2 estimates for 1989 show a slight decrease of about 7% compared with 1985, mainly due to the reduction of Inco emissions. The 1971-1989 Ontario total SO_2 emission trend is shown in Figure 2. The general decreasing SO_2 trend is projected to continue until 1994 when the Countdown limit is expected to be achieved. NO_x emission levels in the 1980's appear to be relatively constant. NO_x emissions from vehicles account for about half of the

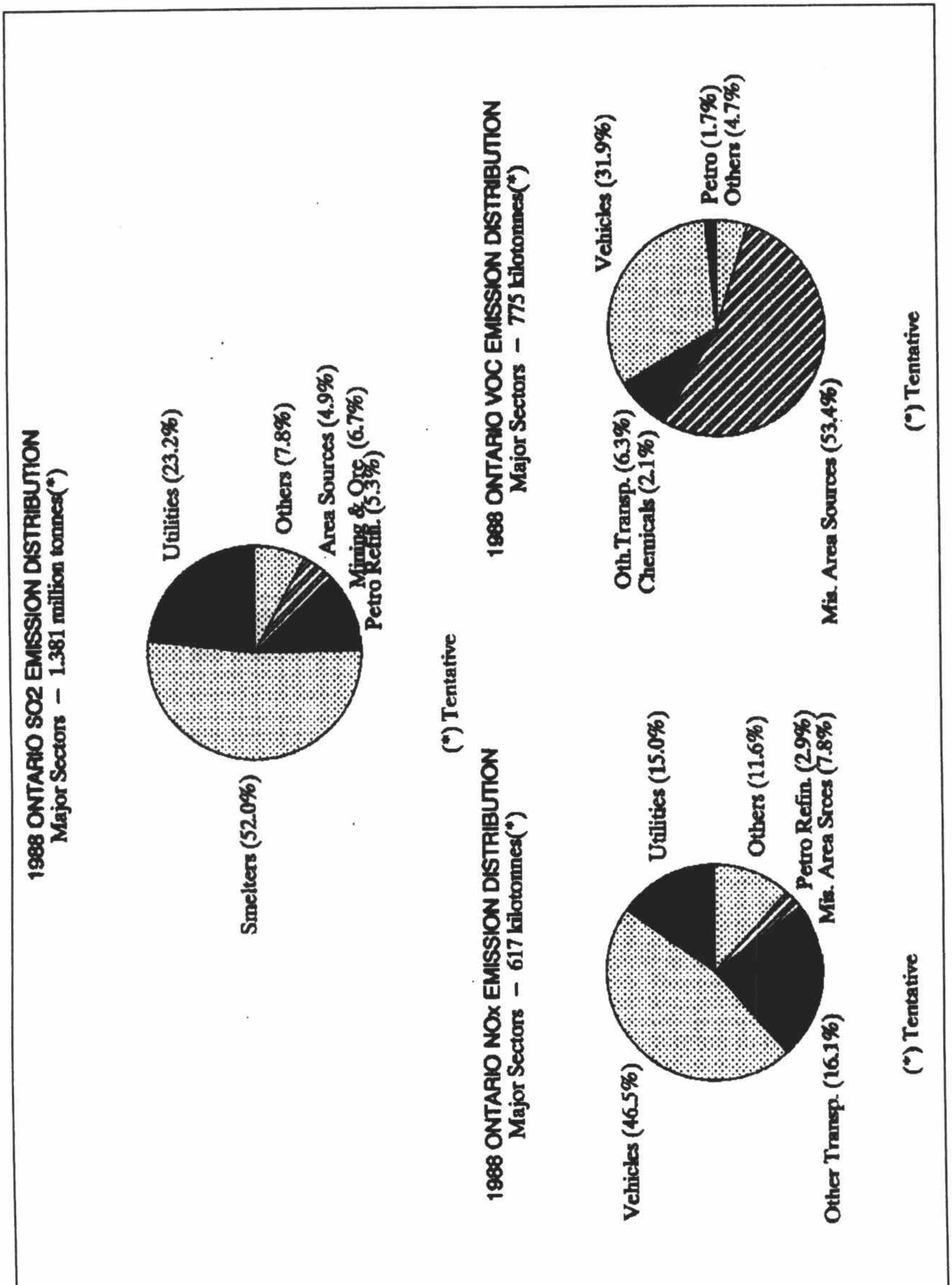

Figure 1 1988 Ontario Base Year Emissions for SO_2, NO_x, VOCs

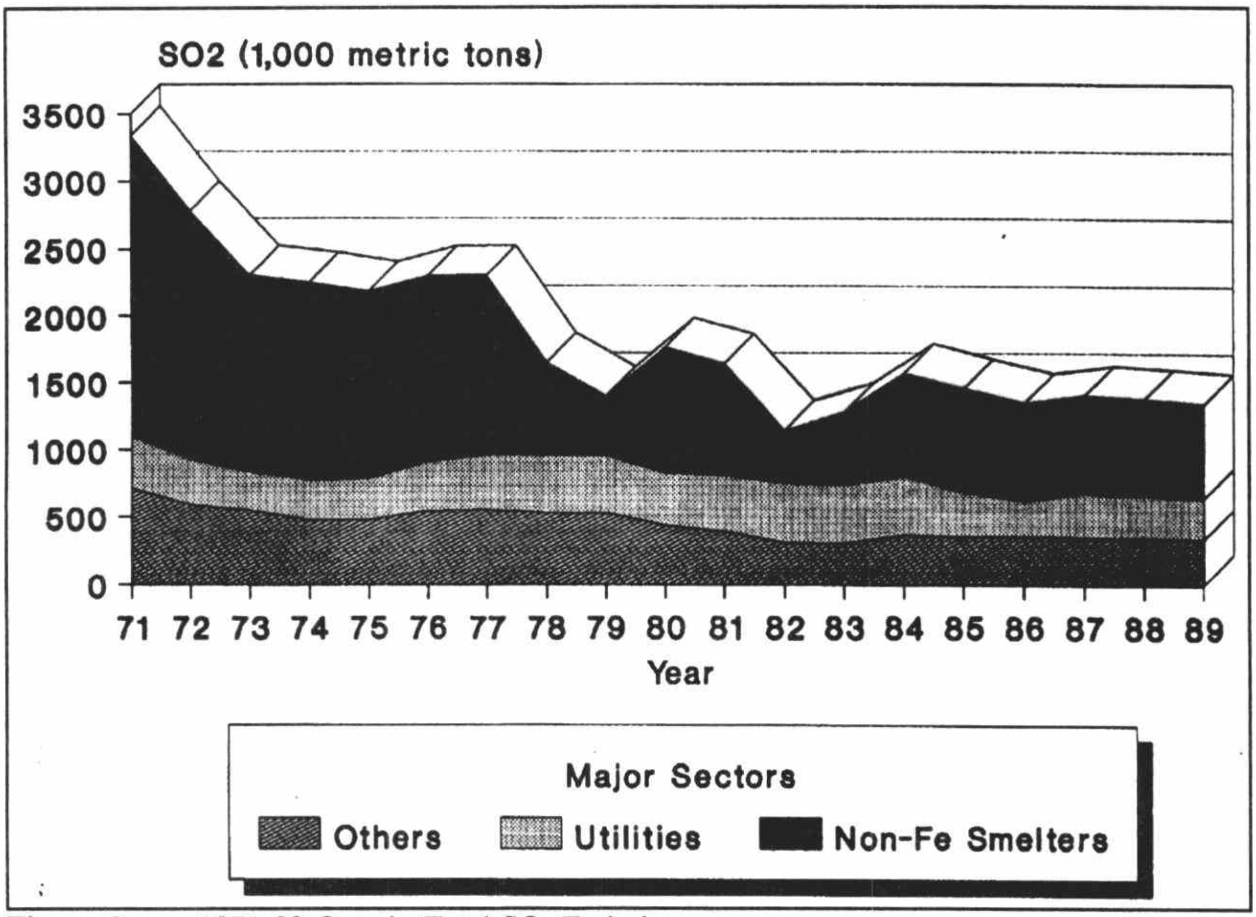

Figure 2 1971-89 Ontario Total SO_2 Emissions

provincial total. With the introduction of more stringent emission standards for light duty vehicles, NO_x emissions are expected to decline in the period to 2000 in spite of the expected increase in vehicle population.

Emission trends of SO_2 and NO_x in North America for the period 1980-1985 were also prepared for the Federal-Provincial Research Monitoring Coordination Committee (RMCC) 1990 Assessment Report during FY 1989/90.

With reference to the global warming issue, a preliminary CO_2 emissions by sectors inventory for Ontario was derived for 1987 (see Figure 3).

B. MODELLING

Numerical models have been used to quantify the source-receptor relationships that exist between acidic precursors emissions and final acid loadings. Linear and comprehensive models have been developed with different applications in mind. Linear models have undergone extensive evaluations (e.g., the Memorandum of Intent between Canada and the U.S. in 1980 and the International Sulphur Deposition Model Evaluation in 1986). Comprehensive models like ADOM (Acidic Deposition and Oxidant Model) have also been evaluated for periods representing

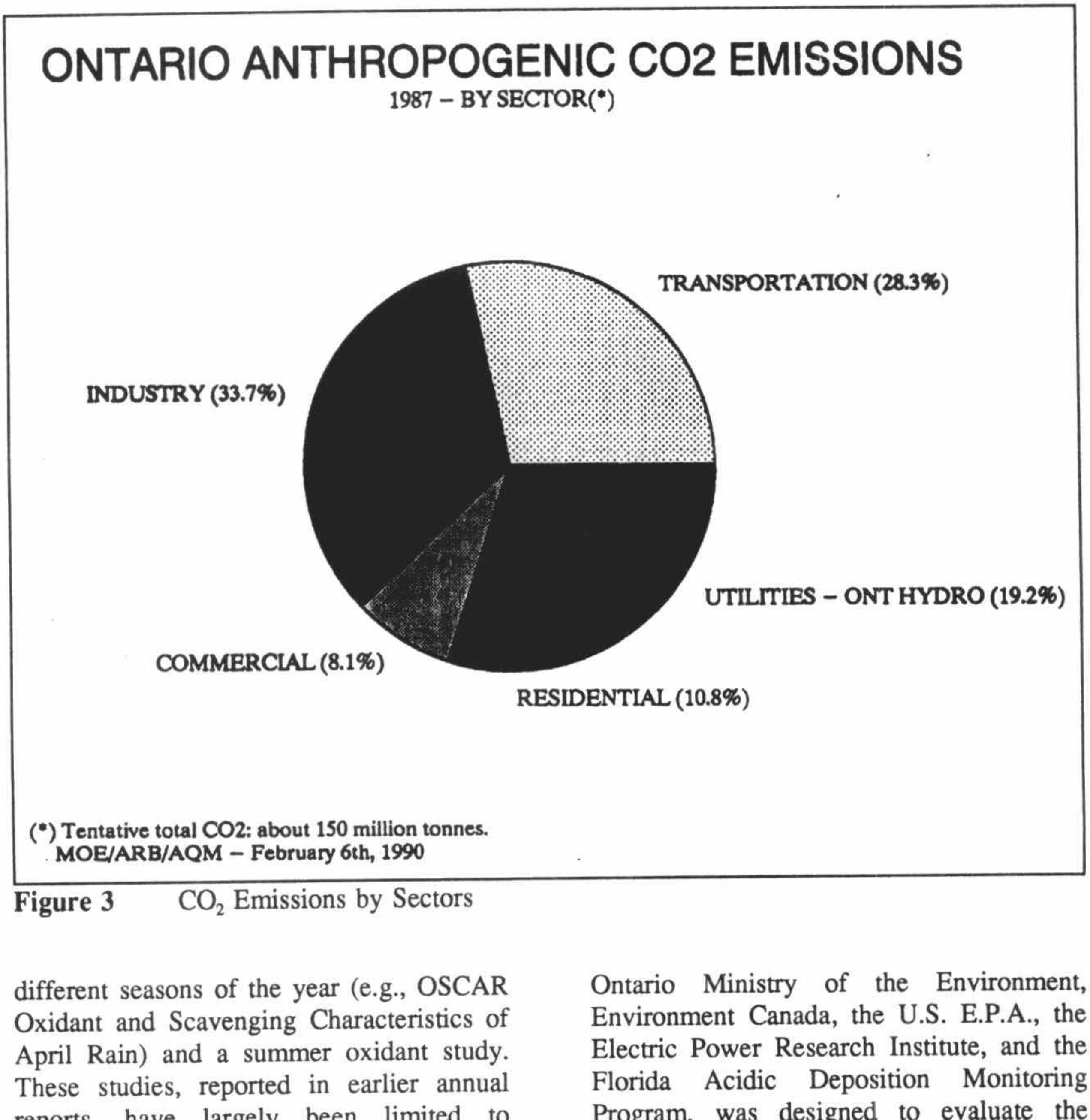

Figure 3 CO_2 Emissions by Sectors

different seasons of the year (e.g., OSCAR Oxidant and Scavenging Characteristics of April Rain) and a summer oxidant study. These studies, reported in earlier annual reports, have largely been limited to comparison of SO_4^{2-} concentrations in precipitation. They show that total deposition of sulphur (wet and dry) is nearly linearly related to SO_2 emissions, and that about 50% of acidic deposition in south-central Ontario comes from U.S. sources. Other modelling studies indicate that nitrogen oxide emission control will result in a decrease in acidic deposition and oxidant levels in southern Ontario. The Eulerian Model Evaluation Field Study (EMEFS), which is a cooperative venture between the Ontario Ministry of the Environment, Environment Canada, the U.S. E.P.A., the Electric Power Research Institute, and the Florida Acidic Deposition Monitoring Program, was designed to evaluate the performance of modern models of long range transport, chemical transformation and deposition. The field study ended on May 31, 1990. An initial model evaluation has already taken place; a more detailed evaluation will be carried out when all of the monitoring data are available.

The first phase of the EMEFS's intensive data collection was conducted between July and September, 1988 and provided both surface and aircraft measurements of air and

precipitation concentrations of a number of key acidic species and oxidants. ADOM was run for parts of the EMEFS period and evaluated during FY 89/90.

During the evaluation process, the model was used in the capacity of a diagnostic tool to identify problems in the NO_x emissions which were common to both ADOM and the U.S. EPA comprehensive Regional Acidic Deposition Model (RADM). This was later confirmed by further investigations. The results of the evaluations for two periods (July 28 - August 8, 1988 (Period 1) and August 25 - September 5, 1988 (Period 2)) are summarized below.

Eight species from the ADOM model have been evaluated. They are the ground level concentrations of SO_2, SO_4^{2-}, total sulphur (SO_2 and particulate SO_4^{2-}), NO_2, nitrate and O_3, and concentrations of SO_4^{-2} and nitrate in precipitation. The performance of the model in predicting these species was as follows:

Ground level SO_2:
Mean overprediction by 10 to 30 percent with higher overprediction at the low end. The model explains about 55 per cent of the variance.

Ground level SO_4^{2-}:
Mean underprediction of 40 to 60 per cent. The model explains between 65 and 85 per cent of the variance.

Ground level Sulphur:
Mean modelled concentration is within 8 per cent of the observed mean.

SO_4^{2-} Concentration in Precipitation:
Mean precipitation-weighted model concentration agrees with observations to within 10 per cent. The model explains less than 16 per cent of the observed variance.

Ground Level NO_2:
Mean underprediction of 5 to 20 per cent. The model explains between 5 to 47 per cent of the observed variance.

Ground Level Nitrate:
Mean underprediction of less than 10 per cent. The model explains about 50 per cent of the variance (Figure 4).

Nitrate Concentration in Precipitation:
Mean underprediction of 20 to 25 per cent. The model explains less than 10 per cent of the observed variance (Figure 5).

Ground level O_3:
Underpredicts the peaks in heavy emissions areas with a slight overprediction of the magnitude of peaks in remote areas.

Preliminary results indicate that the inclusion of non-precipitating stratus clouds in the model improves the results significantly (see Figure 6 for ground level SO_2, Figure 7 for ground level sulphate and Figure 8 for SO_4^{2-} concentrations in precipitation).

The large scatter in the concentrations in precipitation is caused by the mismatch in the spatial scale of representation: the observation is a point measurement which is subject to sporadic precipitation whereas modelled output is a grid average for 127 x 127 km^2. This variability can be reduced by taking longer time averages.

The overprediction of SO_2 in air and the underprediction of SO_4^{2-} in air is common to both ADOM and RADM and it is the focus of current investigations.

A winter simulation was also performed from January 28 to February 7, 1985. The predicted nitrate/sulphate ratio in precipitation was 1.6 which was higher than the predicted spring ratio (during OSCAR) of 0.8. This agrees with the trend in

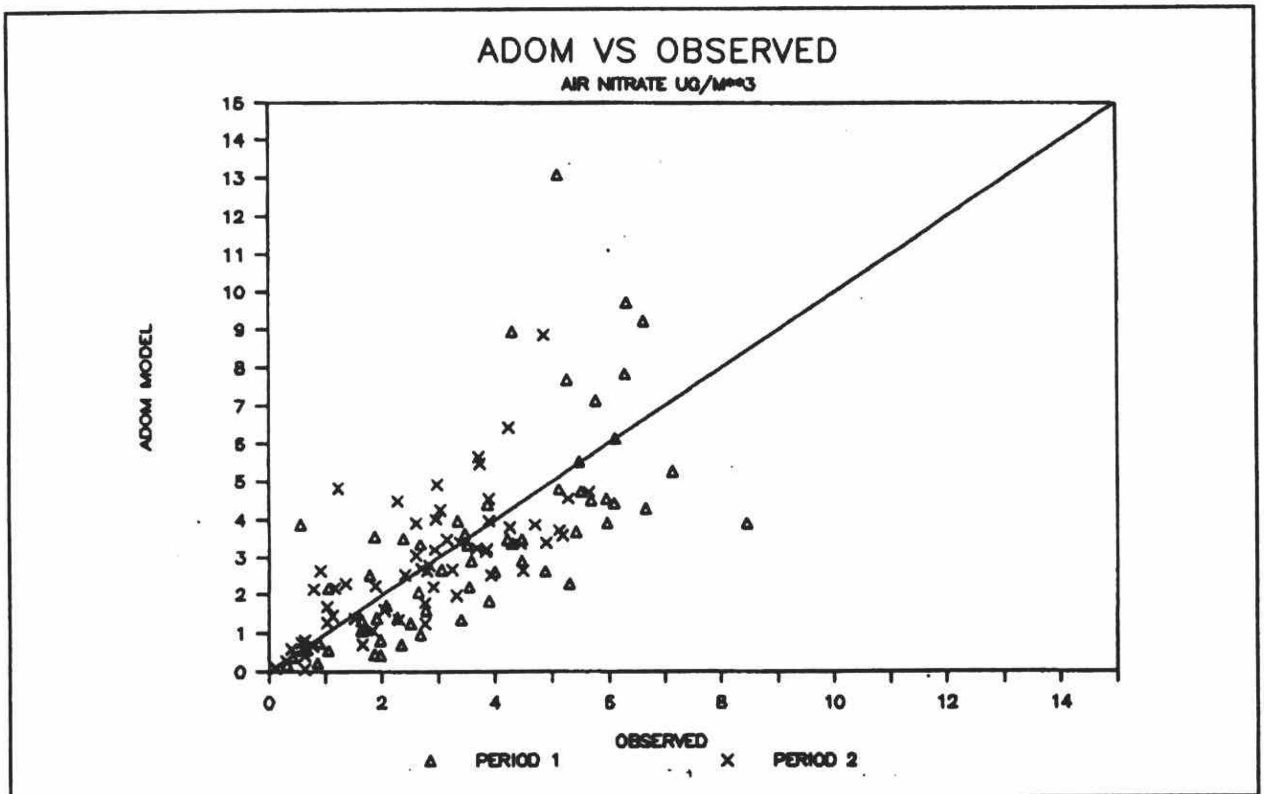

Figure 4 Modelled vs Observed Concentration of Total Nitrate in Air

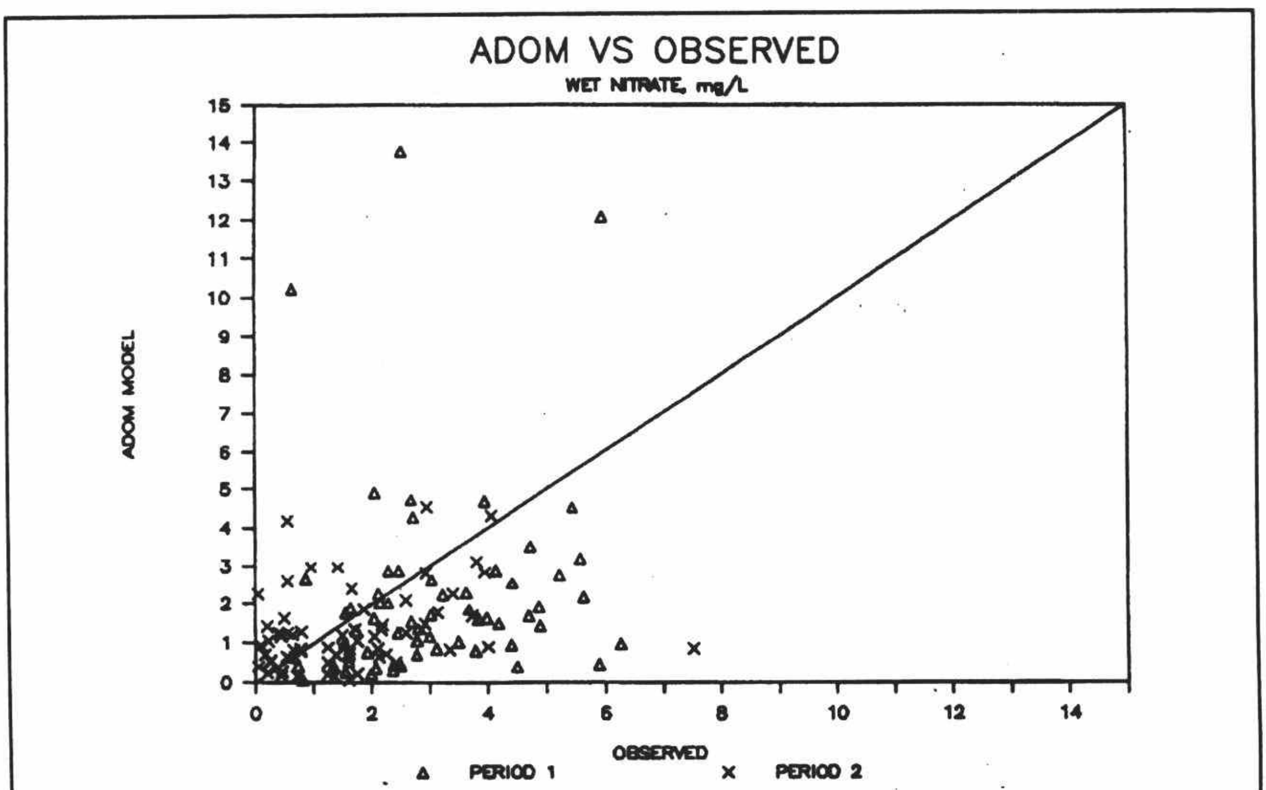

Figure 5 Modelled vs Observed Concentration of Total Nitrate in Precipitation

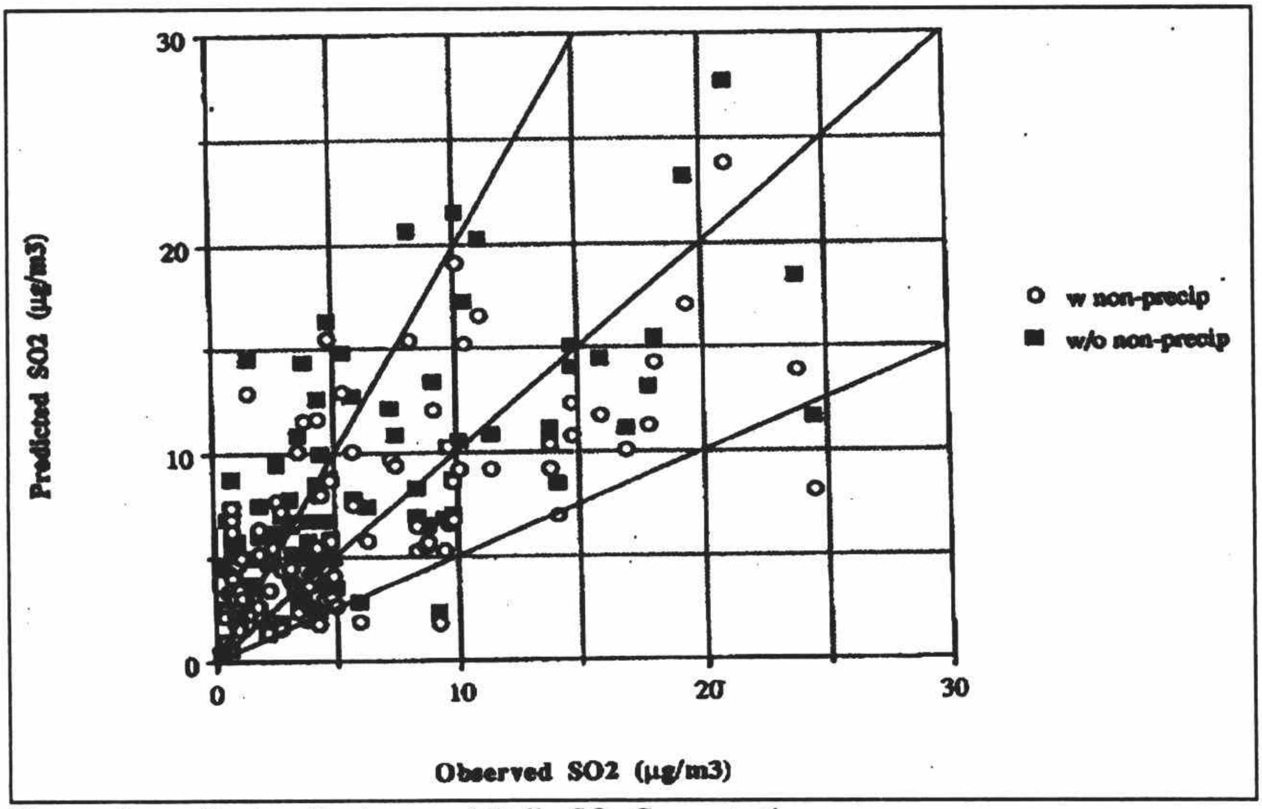

Figure 6 Regionally Averaged Daily SO_2 Concentrations

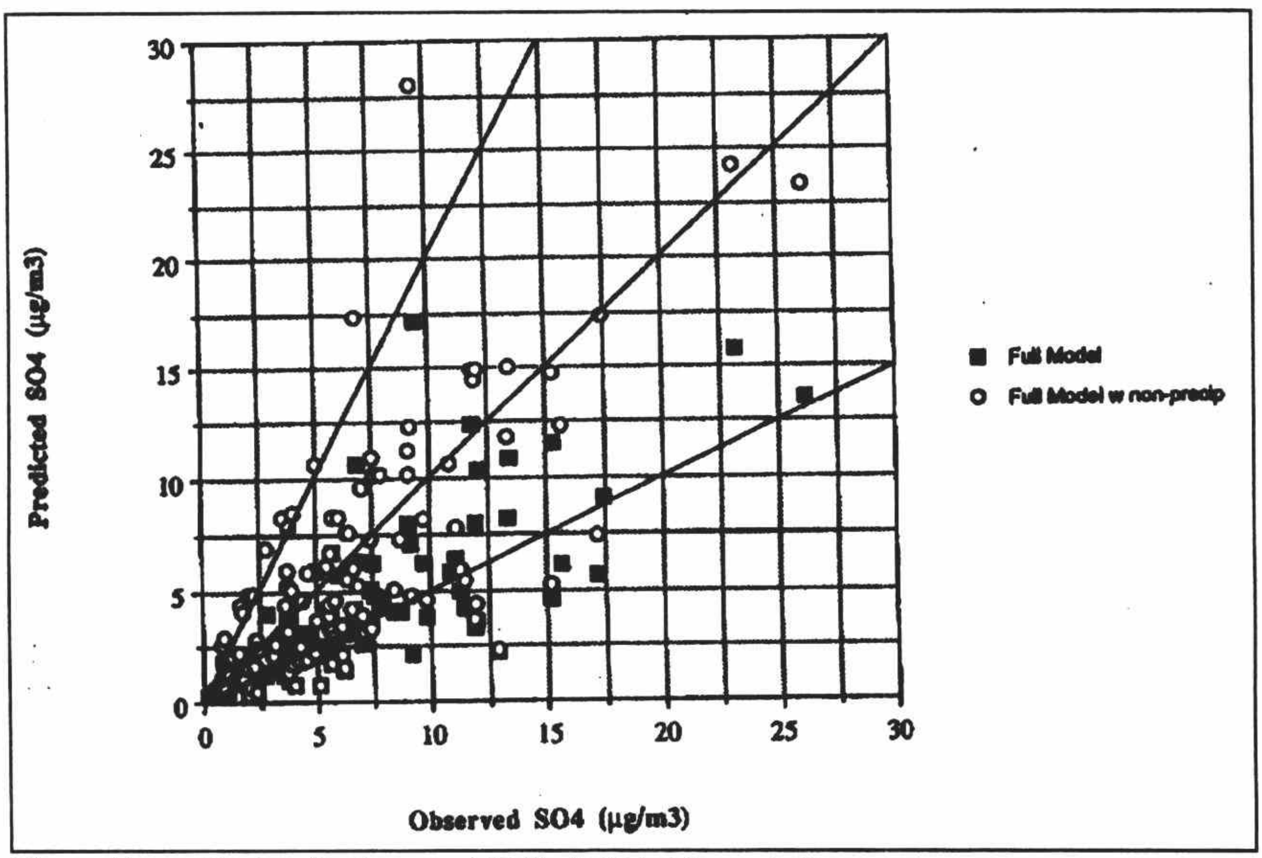

Figure 7 Regionally Averaged Daily Sulfate Concentrations

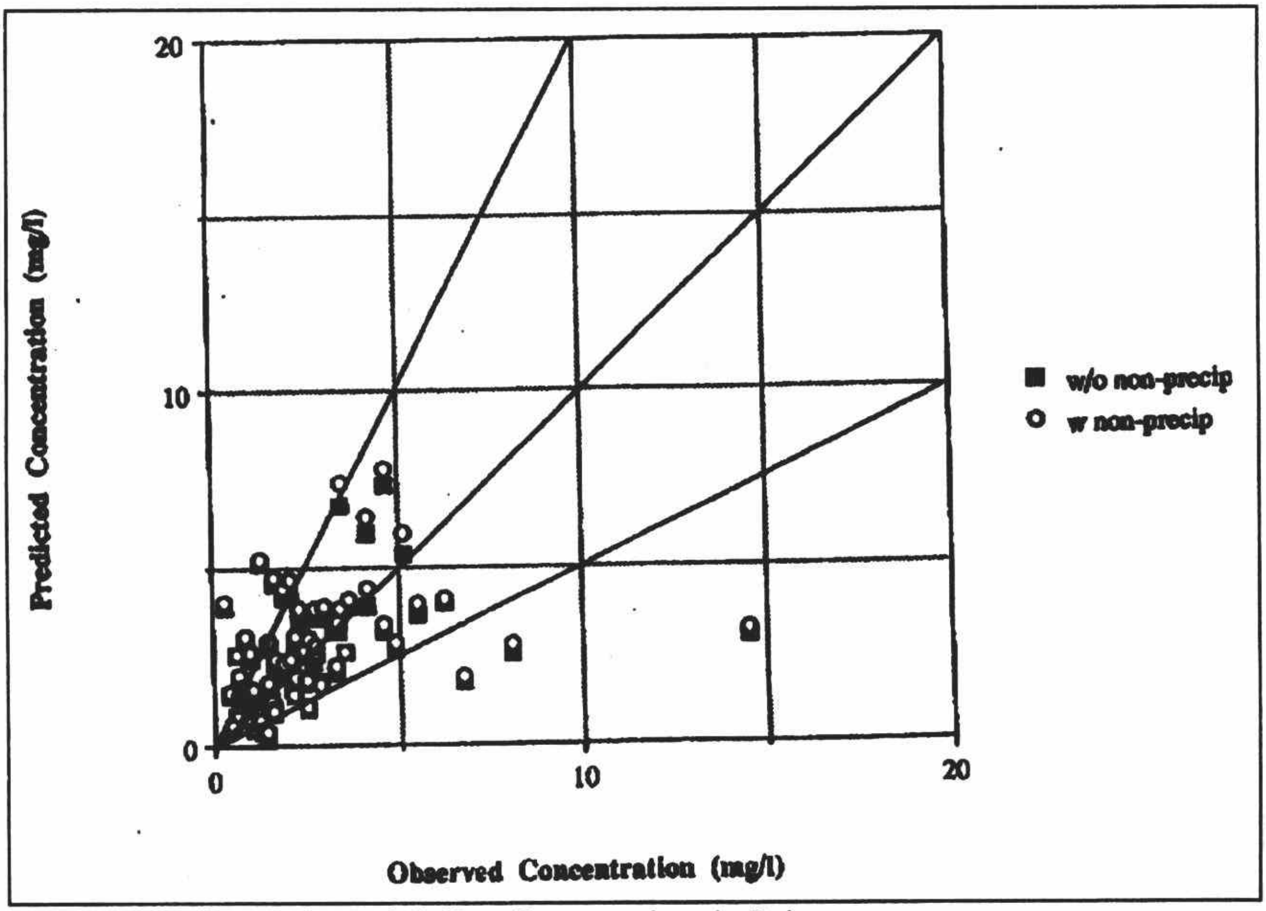

Figure 8 Comparison of Sulfate Concentrations in Rain

observed ratios at the same sites (2.4 in winter and 0.9 in spring). This study investigated the various pathways involved in the production of nitrate that result in the higher winter nitrate/sulphate ratios.

The above modelling works are also contained in the document prepared for the RMCC 1990 Assessment Report during FY 89/90.

A modelling project recently initiated is to adapt ADOM to study the transport, transformation and deposition of mercury. The mercury system (Hg°, HgCl and particulate mercury) has been incorporated into this framework. The sources of mercury in the modelling domain are anthropogenic emissions, natural emissions and elemental mercury (Hg°) residing in the free troposphere which is transported into the domain at the boundaries. The scavenging of the mercury species within clouds involves nucleation scavenging of particulates and continuous exchange of gases at the air - water droplet interface. Observations indicate that more than 95 per cent of the airborne mercury is Hg° which is very insoluble. For this system the determining step in the net scavenging of mercury in clouds is the rate of aqueous phase oxidation of elemental mercury and the original partitioning of the emissions between Hg°, HgCl and particulate mercury.

This model can be used to address a number of questions about the poorly understood mercury system:

- the importance of natural emissions;
- the background air concentrations of Hg°;

- the factors which control the wet deposition of mercury (i.e. possibly O_3 air concentrations and the pH of cloud water droplets).

Preliminary runs have been made to show that the model results are in the range of observed values. Based on new information on emissions and the deposition and scavenging processes occurring, the model will be modified and run for scenarios such as those discussed above.

Another project initiated during FY 1989/90 was to obtain a subgrid scale version of ADOM for use in urban modelling of oxidants and for investigating the spatial variability of acid species.

C. DEPOSITION MONITORING NETWORK

Operation of both the daily and cumulative (28-day) networks continued through FY 1989/90. Data from selected sites in the daily network are included in the Eulerian Model Evaluation Field Study (EMEFS).

Two periods of intensive measurement were embedded within EMEFS. The first took place in the summer of 1988, and was reported previously. The second was carried out during March and April of 1990. The concentrations of a number of chemical species, such as formaldehyde, hydrogen peroxide, ozone, peroxyacetyl nitrate (PAN), etc., were monitored on an hourly time scale at three Ontario locations, including the APIOS Dorset site. Similar measurements were also made on board an aircraft flying between these sites. Measurements made during this intensive study will supplement the network data in model evaluation. In particular they will provide a detailed testing of the model's chemical simulation under early spring conditions.

The cumulative network is the Province's instrument for determining the long term trends in acidic deposition. A recent analysis of data from the Dorset site indicated that sulphate in air and precipitation has decreased since the start of the network in 1980 (Figures 9 and 10), but that nitrate in air and precipitation has not changed appreciably (Figures 11 and 12). These findings, which are in accord with emissions inventory data, have been included in the recent RMCC Assessment Report "Source-Receptor Relationships: Current Understanding and Implications Regarding Emission Controls".

In general, annual wet sulphate deposition exceeds 20 kg/ha/yr over most of southern/central Ontario. There has been, however, a decline in the spatial extent over which the 20 kg target loading is exceeded.

Other key findings remain valid i.e., that nitrogen oxides contribute almost as much to acidic deposition as sulphur oxides (wet and dry), that sulphates dominate in summer precipitation and nitrates in winter precipitation and that the measurable decreases in atmospheric sulphur deposition occurred in response to SO_2 emission changes.

Monitoring of acidic deposition in Ontario will continue for the foreseeable future, so that the effectiveness of controls introduced under Countdown Acid Rain, and elsewhere, can be monitored. However, it is also clear that airborne toxic materials (e.g. pesticides, PCBs, trace metals) are also a significant concern. Rationalization of acidic deposition monitoring has made it possible to monitor these toxic materials without requiring additional resources. To this end, both the daily and cumulative networks have been

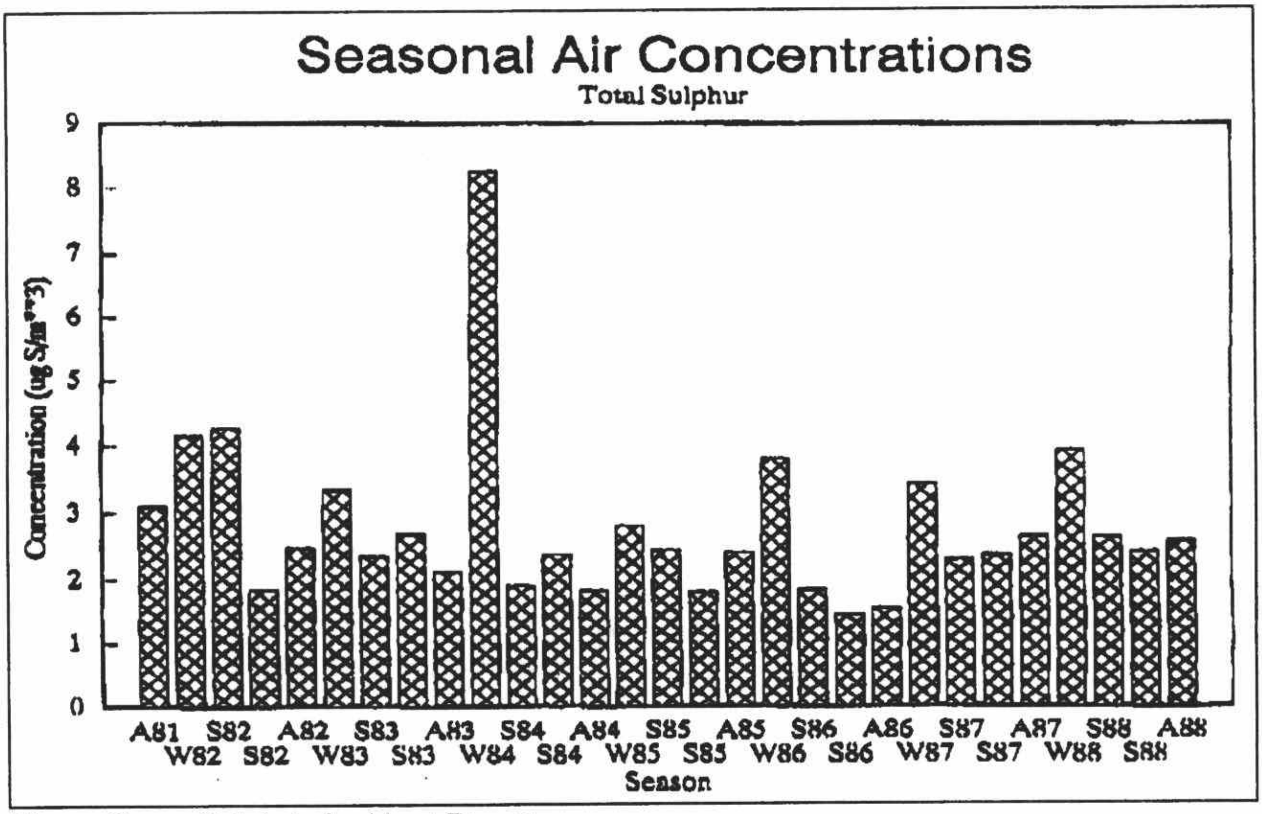

Figure 9 Sulphate in Air at Dorset

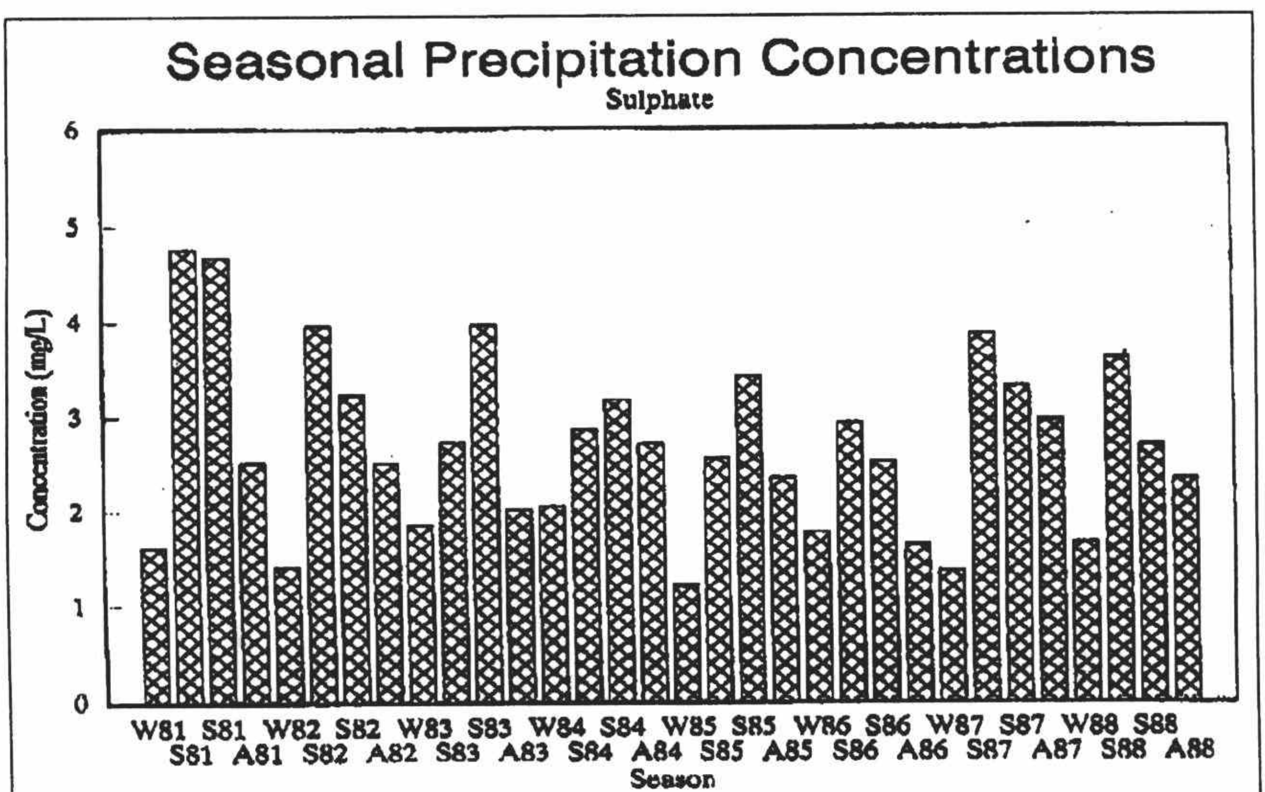

Figure 10 Sulphate in Precipitation at Dorset

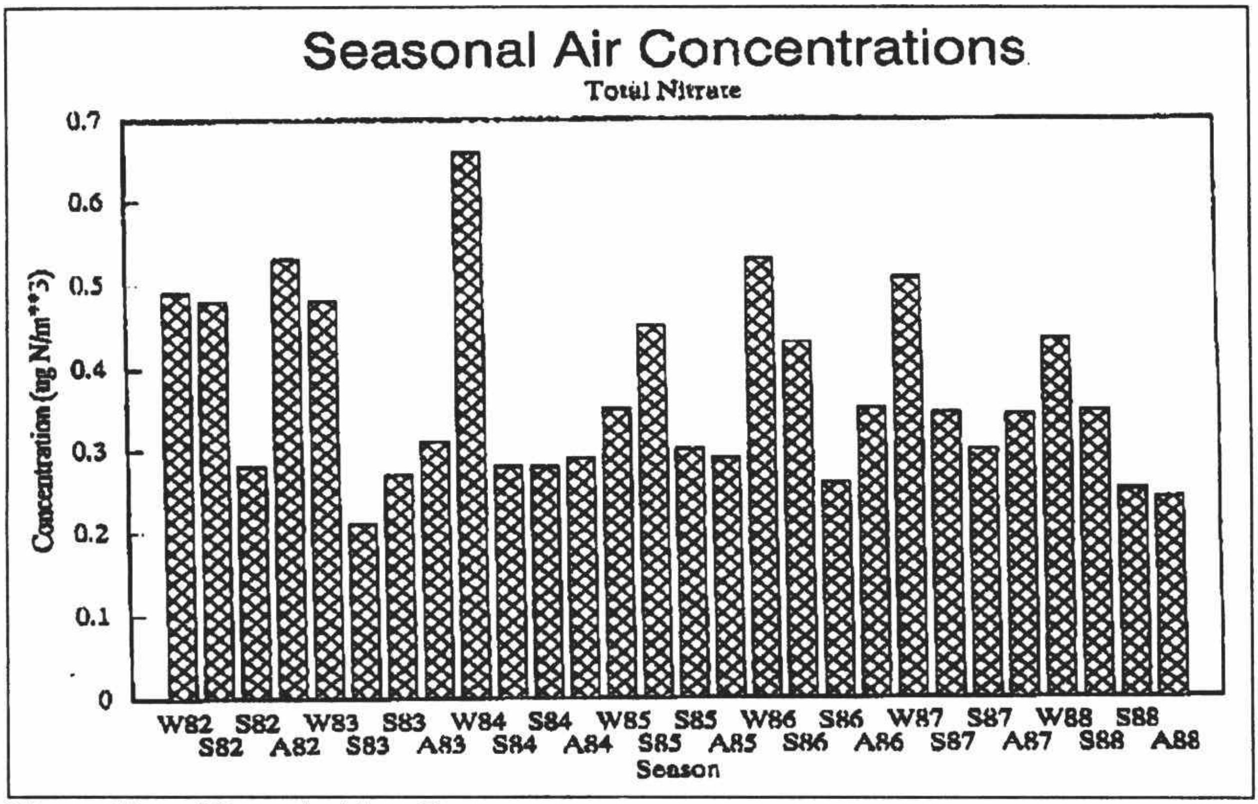

Figure 11 Nitrate in Air at Dorset

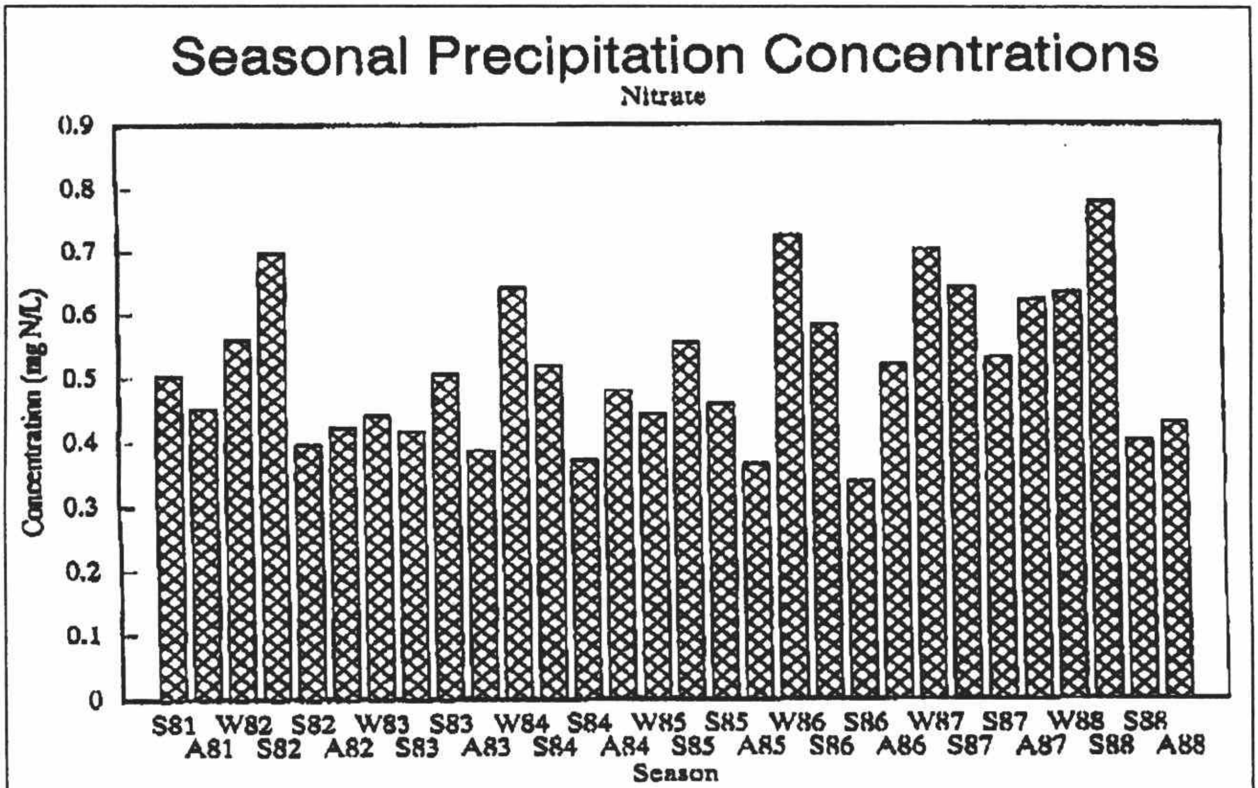

Figure 12 Nitrate in Precipitation at Dorset

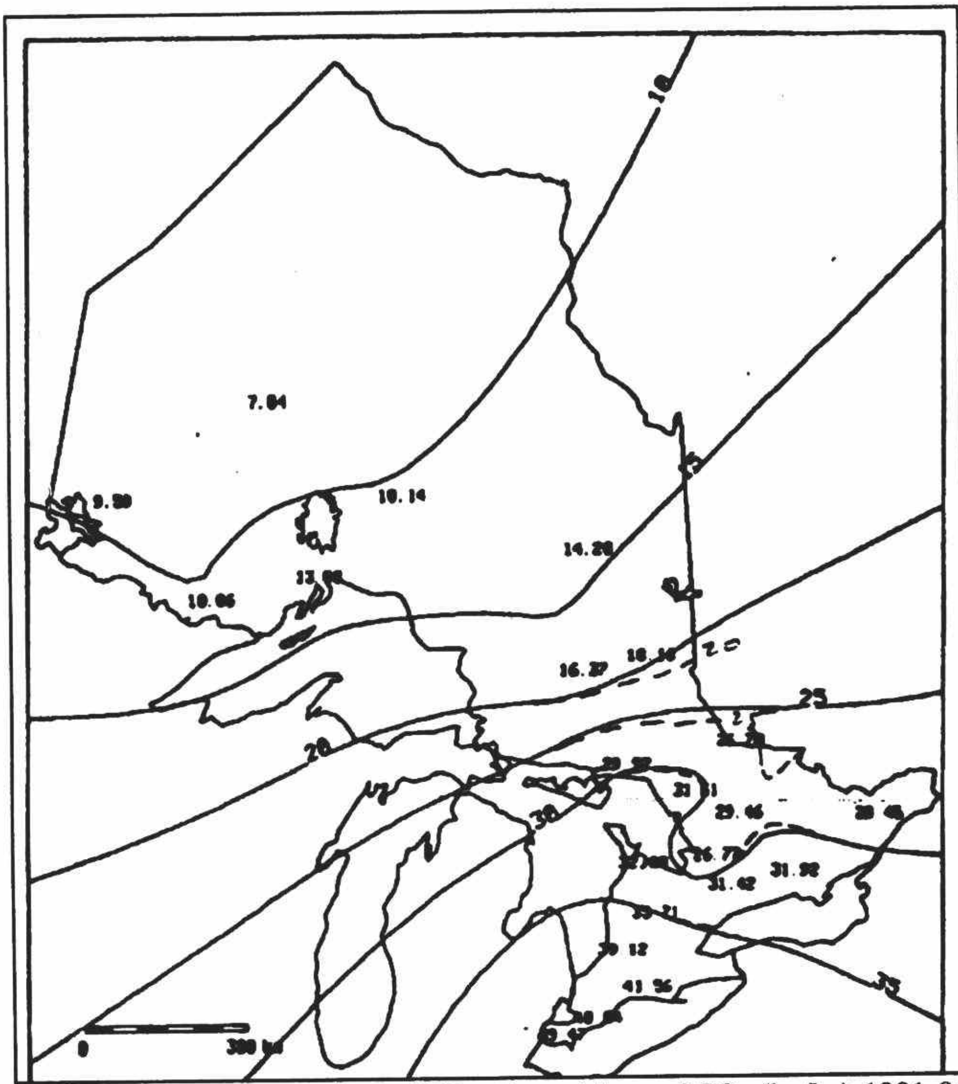

Figure 13 Average Annual Wet Depositions of SO_4 (kg/ha) 1981-86

reduced by the removal of approximately 30% of the sites. Site selection for removal was based on an objective analysis, so that data quality will not be compromised. As an example, Figure 13 illustrates the sulphate deposition pattern calculated for the province with all sites included, and with the selected sites removed.

D. OXIDANTS STRATEGY DEVELOPMENT

An oxidant control strategy document was completed during FY 89/90. The report supports the Ontario initiatives already being considered to reduce oxidant precursor emissions (NO_x and VOCs). These initiatives are the Clean Air Program (CAP) and the Vehicle-related Emissions Strategy. CAP is aimed at the control of all stationary source emissions, including both NO_x and VOCs, as well as air toxics, and will address emission reduction through the application of control technology for the protection of the air environment. The vehicle-related emissions strategy will reduce evaporative emissions through lowering of summer gasoline volatility and through the control of emissions during the distribution of vehicle fuels. Reductions will also be achieved through an active inspection and maintenance program. As an integral part of the program, Ontario supports the Federal initiative to adopt the 1993 California emission standards. Other initiatives, under development cooperatively with other Ministries, include: alternate fuels, changing transportation demand and improved efficiency.

In addition to the above, Environment Ontario has been an active participant in all phases of the NO_x/VOCs Management Plan developed by the Long-Range Transport of

Air Pollutants (LRTAP) Steering Committee for the Canadian Council of Ministers of the Environment (CCME). The purpose of the plan is to address serious health and environmental concerns related to ground-level ozone, NO_x and VOCs on a regional basis through reduction initiatives such as source control, energy conservation, product control, consumer choice and lifestyle and other mechanisms. The final report is expected by November 1990. In support of this and other related actions, regional and urban oxidant modelling studies will be carried out during the next fiscal year.

E. METEOROLOGICAL STUDIES

An operational Meteorological Data Acquisition System (MDAS) has provided on-going support for special studies, episode analyses and modelling activities over the 5-year period 1986 to 1990. MDAS is a computerized system which collects and stores meteorological data supplied by Environment Canada from the North American network of weather stations. Air parcel trajectories are generated by the system for interpreting event precipitation and other air quality data.

During FY 1989/90, a satellite dish was acquired and installed as part of the Anikom 100 service to receive meteorological data from Environment Canada. The data acquisition via satellite transmission was successfully implemented and became operational in 1989. With increased data and data transmission rate, an improved MDAS computer system is required. Accordingly, feasibility studies are currently underway to enhance the MDAS system and to replace the aging Eclipse S/130.

As part of the RMCC 1990 Assessment Report, a study on ozone in southern Ontario for the period 1979 to 1988 was prepared. Data analysis indicates no apparent long-term trend in ground level ozone concentrations. Meteorological analysis of the ozone data clearly shows the importance of emission sources in the United States to ozone air quality standard violation in southern Ontario. Other related meteorological data analyses during FY 89/90 include ozone episodes and the air pollution index.

The sector analyses study of the relative contribution of emissions in the U.S. and Canada to acidic wet deposition in Ontario for the period 1981 to 1985 was completed. The results show the U.S. sector as the dominant contributor to the total annual wet deposition of H^+, NO_3^- and SO_4^{2-} in the border regions of Ontario, typically 80% for southwestern Ontario and 60% for southeastern Ontario. Over this five year period, the decreasing trend in concentrations of the acidifying pollutants in precipitation appears to be linked to changes in regional SO_2 emissions.

Aquatic Effects Studies (Task 2)

Contact: W. Keller

The Aquatic Effects Studies are divided into three major subtasks; Chemical Studies, Biological Studies and Remedial Methodologies Development.

A. CHEMICAL STUDIES

Calibrated Watersheds

a. Trends in Deposition and Lake/Stream Chemistry

In response to a decrease in SO_2 emissions in eastern North America in the last decade, the atmospheric deposition rates of SO_4^{2-} and H^+ have decreased by 35% to 40% in central Ontario. Averaged over the period 1976-85, the mean change in the annual average SO_4^{2-} concentration in precipitation was about 4 $\mu eq\ L^{-1}\ yr^{-1}$. The decline in H^+ concentration was 3.3 $\mu eq\ L^{-1}\ yr^{-1}$ while nitrogen species and base cation concentrations remained unchanged.

During this time, however, Plastic Lake, in the Muskoka-Haliburton region of Ontario, which has been monitored since 1979, acidified. Its alkalinity dropped an average of 2 $\mu eq\ L^{-1} yr^{-1}$ between 1979 and 1985. For the same period, total base cations decreased in the lake while SO_4^{2-} did not change significantly. Since 1986, however, sulphur (and acid) deposition at Dorset has not declined further. The lake's alkalinity and pH, correspondingly, have not continued to decrease.

The chemistry of the streams draining the Plastic Lake catchment has also been monitored. Water quality of the runoff in an upland site improved rapidly in response to decreased SO_4^{2-} and H^+ deposition; pH and alkalinity increased, while SO_4^{2-} and Al decreased. These improvements, however, were negated by a small wetland area downstream and the extremely dry summers of 1983, 1987 and 1988. The wetland reversed most of the changes in water quality (also see Biogeochemistry Studies Section) and the dry periods resulted in very high concentrations of SO_4^{2-} in the streamwater, probably due to re-oxidation of substantial amounts of reduced sulphur in the catchment.

Sulphur emissions from Sudbury also declined between 1978 and 1985 by an estimated average of 0.73 X 10^6 tonne yr^{-1}. As a result, SO_4^{2-} concentrations of lakes in the Sudbury area have decreased and the pH of each of the acidic lakes studied has increased. Aluminum, Cu, Ni, and Zn concentrations have also decreased, likely as a result of decreased emissions from the smelters.

These results, together with those from the upland site at Plastic Lake, clearly demonstrate that chemical acidification of aquatic ecosystems is reversible, and that beneficial effects from the reduction of sulphur deposition can be expected within a few years to a decade of the deposition decreases.

b. Modelling Lake Chemistry

A better understanding of the acidification mechanisms can be obtained by a combination of field studies and mathematical modelling. A complete model would require very extensive information about the catchment (e.g., precipitation, water chemistry, soil chemistry, vegetation). Few data sets satisfy this requirement; however, a modelling effort is still worthwhile and may provide the insight required to plan further field work.

The Birkenes model, previously used for modelling stream chemistry in Norwegian catchments was modified to predict discharge and sulphur concentration in a stream draining the Harp Lake catchment in Ontario.

The predicted hydrology is generally in close agreement with the measured hydrology. Good agreement between calculated and observed sulphur concentrations was obtained when a reduction (or adsorption) process for SO_4^{2-} in the deeper soil horizons was introduced. During dry periods, however, SO_4^{2-} is produced by oxidation in the upper soil layer. Sulphate concentrations in the stream water during snowmelt are remarkably constant and, according to the model, could be explained by the existence of an easily soluble sulphur reservoir in the soil, which is considerably larger than the annual sulphur flux.

Even with the modifications for Harp Lake, the basic structure of the models applied in Norway and Ontario was the same, and it seemed likely that the model would be applicable to other catchments with only minor modifications. Application of the model to two other subcatchments in the Harp Lake watershed proved successful.

The model was then extended to include a hydrological submodel with a snow reservoir and two soil reservoirs, a sulphate submodel and an ion submodel which included H^+, Ca^{2+} + Mg^{2+}, Al, Na^+, HCO_3^- and organic anions. Major processes incorporated included different hydrologic flow patterns during highflow and lowflow, SO_4^{2-} adsorption and desorption, ion exchange, weathering, gibbsite (amorphous alumino-silicate) solubility and CO_2 equilibria in soil and stream water. The model reproduced important daily and seasonal trends in the observed streamwater chemistry of a tributary to Harp Lake.

Finally, this extended model was run to simulate a doubling and a 50% reduction in the 1983 sulphur deposition. The simulations showed little change in lowflow stream pH with changing deposition. At snowmelt highflow, however, a 50% reduction in deposition caused a pH increase of 0.2 to 0.5 unit. Doubling the deposition resulted in pH depressions of 0.5 to 0.9 unit.

Within the last two years the MAGIC model has also been used. The MAGIC model began as a groundwater model but can now be used to model streams and lakes.

The model consists of a soil solution equilibrium component and a mass balance component. The MAGIC model takes a series of equilibrium and mass balance equations and a collection of parameter definitions and fits modern water and soil chemistry. The model results are strongly driven by historic deposition chemistry.

For Plastic Lake, which has experienced a four-fold decrease in alkalinity and a 0.25 unit drop in pH over the past nine years, the model was successful in predicting historic water chemistry (ANC, SO_4^{2-} and pH). The model, however, did not predict the observed trends in alkalinity and pH. According to the model, decreased sulphur deposition

should have led to rapid reversal in the acidification of Plastic Lake. Poor prediction is probably due to sulphur storage in the wetlands which is released over a long period of time, lending a lag time to the recovery of the lake.

c. Effects of Nitrogen

The relative contribution of HNO_3 to precipitation acidity in eastern Canada has increased in recent years. This caused concern that the relative importance of NO_3^- deposition in the acidification of terrestrial and aquatic ecosystems may increase.

Alkalinity and pH depressions in streams and lakes, as a result of snowmelt, are seen in the Muskoka - Haliburton area, and have been well documented in the literature over a broad geographical scale and a wide range of alkalinity. It was thought that in areas of high acid deposition SO_4^{2-} and NO_3^- may dominate these alkalinity depressions when anions, accumulated in the winter snowpack, are released over a brief melt period. The relative contribution of base cations and acid anions to alkalinity decreases was measured on 15 headwater streams and lake outflows in three central Ontario catchments. It was found that neither nitrate nor organic acids were significant contributors to the observed alkalinity depressions. Base cation dilution by snow meltwater was found to be the major contributor to alkalinity depressions. Cations are very low in the snowpack and very little is picked up from the soil.

Further studies on the importance of nitrogen included the calculation of annual mass balances for NO_3^- and NH_4^+ for several forested catchments and lakes. Ammonium retention by forested catchments was consistently high compared to NO_3^- retention. Inorganic N retention was influenced by catchment grade and areal water discharge, where areal water discharge equals the ratio of lake mean depth to water residence time.

In the lakes, NO_3^- and NH_4^+ export were linearly related to areal water discharge. Nitrate retention did not seem to be a function of the degree of acidification of the lakes. Nitrogen consumption-related acidification was most likely to occur when the areal discharge was less than 1.5 m yr^{-1}.

d. Mass Balances

Alkalinity

It recently has become apparent that internal (in-lake) processes are significant sources of alkalinity, and in some cases more important than external sources such as silicate and carbonate dissolution, ion exchange reactions and redox processes.

Alkalinity and ion budgets were measured for eight lakes and 24 subcatchments in southern Ontario for periods ranging from 6 to 10 years. The contributions from the lakes' catchments and from the atmosphere were quantified.

For Harp and Plastic Lakes, which have the highest and lowest alkalinities, respectively, the loss of alkalinity from each lake via the outflow greatly exceeded the total input. In Plastic Lake there was an output of alkalinity even though the input was negative.

If the catchment were the only source of alkalinity in each case (external source) the alkalinity concentration in Plastic Lake should be -75 $\mu eq\ L^{-1}$ and in Harp Lake should be 13 $\mu eq\ L^{-1}$. In fact, the measured alkalinities are 8 and 64 $\mu eq\ L^{-1}$ respectively, indicating a significant contribution from in-lake processes. Mass balance calculations indicated that the major in-lake contributors were the removal of organic anions followed

by SO_4^{2-} and NO_3^- reduction.

Cations

Cation flux from the lakes' catchments was found to be controlled by the atmospheric input of strong acids of anthropogenic origin. Organic acids and H_2CO_3 were less important factors.

Sulphate

Over the past 10 to 15 years there has been a decrease in the atmospheric deposition of SO_4^{2-} and strong acid as a result of decreased SO_2 emissions. The SO_4^{2-} budgets for the Plastic and Harp Lake catchments indicate that the export of SO_4^{2-} from both catchments exceeds the input on an annual basis over several years. A new steady state will eventually be reached.

On a short-term basis the SO_4^{2-} fluxes are not in equilibrium. In the summer, SO_4^{2-} is stored in the catchment. In the winter and spring, there is a net release of SO_4^{2-} The storage and release are almost certainly controlled by redox processes. In very dry summers, streams dry up and oxidation of the stored sulphur occurs. On return of stream flow, flushing of H_2SO_4 from the catchment results in extreme conditions that are as detrimental as, or worse than, the spring snowmelt.

Carbon

To better understand the cycling of dissolved organic carbon (DOC) in the watershed, carbon isotope analysis (^{13}C and ^{14}C) was done on the DOC and dissolved inorganic carbon (DIC) in Harp Lake.

At the study site soil gas, lysimeter leachate, piezometer and bedrock well water, stream, precipitation and sediment samples were collected. DOC concentrations were compared for the period of August 1988 (low flow) and April 1988 (high flow).

The major findings of this work can be summarized as follows: a) the lake is a sink for DOC and therefore organic acids; b) in the summer there is an in-stream source of DOC; c) 50% to 74% of the DOC is hydrophobic (humic and fulvic acids, and neutrals); d) the wetlands and beaver ponds have older carbon; e) a rapidly cycling component exists apart from the fulvic and humic acids; f) the groundwater contains older carbon which suggests a fractionation or cycling of DOC in the upper soil zone; g) all the alkalinity in Harp Lake is from silicates, and h) the lake is exporting CO_2 at all times of the year.

e. Acidification and Phosphorus Export

Acidified lakes are often exceptionally clear, and it has been hypothesized that this phenomenon is the result of a decline in productivity driven by a fall in phosphorus output from acidified watersheds. The chemical theory underlying the hypothesis was tested on streams in the Muskoka-Haliburton region of Ontario.

Watersheds with clear, circumneutral streams will export more phosphorus when further acidified. Coloured, acidic streams already export elevated amounts of phosphorus. Phosphorus loadings to a lake will, therefore, depend on the relative inputs of clear water and brown water streams.

Metal Contaminants

a. Aluminum

The acidified lakes and streams in the Muskoka - Haliburton region have been found to contain high levels of aluminum. In order to determine the sources, sinks and forms of aqueous aluminum, 26 different

streams in the Plastic and Harp lake catchments, and 60 lysimeters at different locations in the catchment and soil profile were monitored.

Inorganic monomeric aluminum was found to originate primarily from the Bf horizon in the soils and was found in all downstream locations. The mineralogical source and sink for inorganic monomeric aluminum appeared to be an aluminum trihydroxide solid phase. Organic monomeric aluminum and dissolved organic carbon were obtained from two widely separated sources: the soil LFH horizon and the wetland.

b. Mercury

Environment Ontario and the Ministry of Natural resources have surveyed contaminants in the sport fish from Ontario lakes since the 1970's. Survey data have revealed that mercury contamination in Ontario fish is common and widespread in lakes on the Precambrian Shield, even though most lakes are remote from industrial or municipal contamination sources.

Mercury levels in the large size classes of fish often exceed the concentrations considered safe for eating. As a result, size specific consumption restrictions have been placed on fish from most of these lakes.

Initially, the mercury in these lakes was thought to be primarily from natural sources (probably geological) since the affected lakes were mainly on the mineralogically rich Precambrian Shield. It was also hypothesized that the mercury distribution in these lakes might be, in part, due to atmospheric deposition. Until recently, however, the analytical capabilities for measuring mercury in water were inadequate to correctly assess the relative importance of these sources.

Within the last five years, the necessary analytical capabilities were developed for the measurement of mercury at low levels in water samples. Runoff from a group of calibrated watersheds and precipitation in the Muskoka/Haliburton area of south-central Ontario were examined. The annual volume weighted concentration of mercury in precipitation was four to eight times higher than in streams. In a typical headwater lake, more than half of the annual mercury load to the lake entered directly through precipitation. Most of the watershed-derived mercury entered the lake during spring runoff.

It is likely that much of the mercury in the runoff was derived from precipitation. Most of the mercury in the watershed was removed from the atmosphere. Mercury export from each watershed was only 10% to 20% of the atmospheric load to the watershed.

Current data, therefore, clearly indicate that contrary to previous thought, the mercury in fish from Precambrian Shield lakes is derived directly from atmospheric deposition and not bedrock weathering.

The retention of mercury deposited on watersheds from the atmosphere varies considerably. The amount exported increases linearly with the amount of wetland in the watershed. Wetlands appear to retain only about 20% of the mercury deposited on them, whereas upland areas retain more than 90%. The variation in retention may explain some of the variation in the mercury content of fish in lakes on the Precambrian Shield.

The relative importance of anthropogenic and natural sources of mercury in the atmosphere is under debate. Mercury concentrations in sediment profiles suggest that, as found in other North American

locations, mercury input to lakes in Ontario has increased two to three fold since pre-industrial times. Scattered measurements across the northern and southern hemispheres indicate a gradient in atmospheric concentration with the most southerly stations exhibiting the lowest concentration. Although emission inventories prepared in the mid-1970's suggest that natural sources of mercury play a larger role than anthropogenic sources, mass balance calculations using the most recent data suggest an overestimation of soil emissions by a factor of 10. Present data indicate that the current mercury load to the atmosphere (and eventually to the lakes) is largely of anthropogenic origin.

c. Other Metals

The trace metals Cu, Pb, Cd and Zn and their patterns of transport within the Plastic Lake catchment in the Muskoka - Haliburton region were also examined. The aqueous metal concentrations were followed from the precipitation through the catchment to a DOC, acidified soil water seep, a high DOC wetland outflow and finally to the Plastic Lake outflow.

Lead and Cu patterns are identical to DOC and Al. Levels are relatively high in precipitation, with their lowest median concentrations occurring in the soil water seep, elevated concentrations in the wetland ouflow and lower concentrations at the lake outflow. In contrast, Cd and Zn have their highest median concentrations in the acidified soil water seep and reduced values further downstream.

Lead and Cu transport patterns can be explained by the metals' affinity for DOC and transport patterns for Cd and Zn can be explained by the metals' solubility in dilute acid and mineral soils.

Extensive Lake Monitoring

For the past six years, Environment Ontario in co-operation with the Ministry of Natural Resources, conducted lakes surveys to determine the sensitivity of Ontario lakes to acidic precipitation. Results have been summarized each year in a report entitled "Acid Sensitivity of Lakes in Ontario" issued by Environment Ontario.

During the last 4 years the emphasis has been on the collection of data on lakes underrepresented in the existing data base. These lakes were smaller lakes (< 10 ha) in all of Ontario, and lakes of any size in northwestern Ontario. The percentage of all lakes sampled in each part of the province, is illustrated in Figure 14.

By examining only low conductivity lakes, quantitative relationships between sulphur deposition and the sulphate content of lake water were established. Similar relationships were developed for lake pH and alkalinity.

A detailed descriptive analysis of the 6,000 lakes in the Acid Sensitivity Database was also completed. Lakes were grouped by quaternary watershed according to zones of sulphur deposition. For several lake size groupings, the available data were extrapolated to the total number of lakes in the sulphur deposition zone.

This process yielded the most accurate estimates to date of the number of lakes with alkalinity <0 and lakes with a pH < 6 in the province (i.e., pH < about 5.0). Over 7,000 lakes were estimated to be acidic, and over 19,000 were estimated to be acidified to less than pH 6, the point at which some biological damage occurs.

The most acidified area of the province is the Sudbury deposition zone, where impacts from the sulphur emissions from the

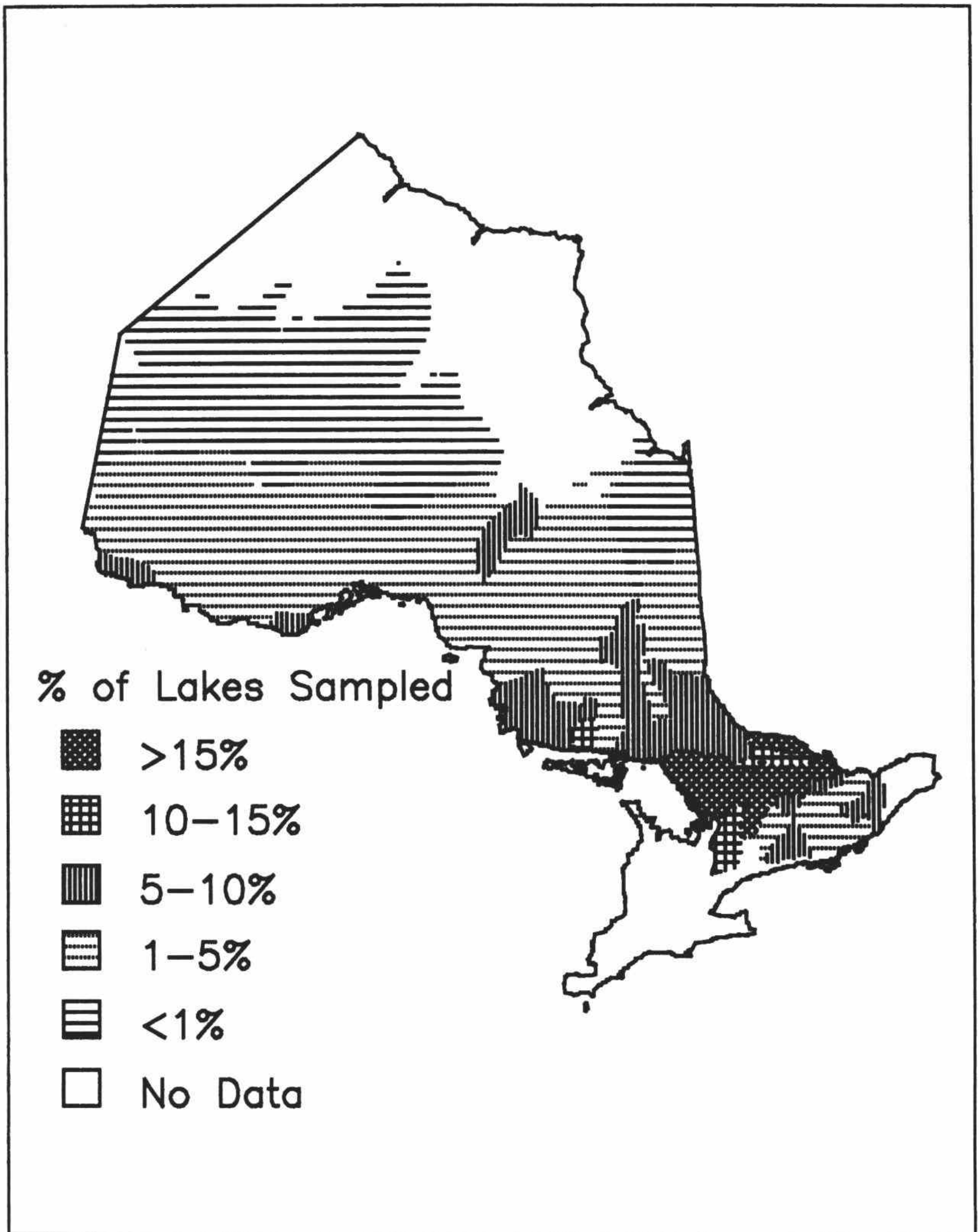

Figure 14: Percentage of Lakes Sampled in Each Part of the Province

Sudbury smelters were greatest. Large numbers of acidified lakes, however, occur throughout central Ontario (Haliburton, Muskoka, Parry Sound, Nipissing, and parts of Algoma and Timiskaming).

Within each of the sulphur deposition zones, the degree of lake acidification was a function of lake size. Smaller lakes are generally more acidic than larger lakes, with lakes in the 1-10 ha range accounting for most of the acidified lakes.

An analysis of the chemical data collected for a total of 1,180 soft-water lakes surveyed between 1981 and 1987 showed a strong relationship between both the mean lake pH and mean lake alkalinity and the sulphur deposition of the zones (Figure 15 and Figure 16).

Assuming that the chemical characteristics in the lowest deposition zone indicate "background conditions", substantial alkalinity has been lost in all of the other regions.

The large sample size of the survey data also allowed it to be used to assess the so-called "critical" load of SO_4^{2-} for Ontario lakes. The statistical relationship between sulphur deposition and lake pH or alkalinity was used, in combination with the defined lake pH or alkalinity below which biological effects occur, to estimate the critical or threshold sulphur deposition below which effects are not observed.

There was a significant relationship between sulphur deposition and lake chemistry only for sensitive lakes (i.e., softwater lakes defined as those with a conductivity of < 50 µs cm^{-1}). This implies that, in lakes with higher conductivity, either SO_4^{2-} deposition does not control lake chemistry or that there are other sources of SO_4^{2-} in these lakes. The critical lake pH of 6.0, corresponding to an alkalinity of about 50 µeq L^{-1}, is generally agreed to be the level below which biological damage occurs. This figure was used to calculate the portion of lakes in Ontario which will not drop below these chemical criteria when exposed to differing levels of SO_4^{2-} deposition.The results, summarized in Table 1, indicate that in order to protect 95% of the sensitive lakes in Ontario, the SO_4^{2-} deposition must not be greater than 9 kg ha^{-1} yr^{-1}, while less than or equal to 15 kg ha^{-1} yr^{-1} will protect 90% of the lakes.

Table 1.
pH Above Which 95% and 90% of Ontario's Sensitive Lakes Will Remain for Specific Sulphate Deposition Rates.

Total SO_4^{2-} kg ha^{-1} yr^{-1}	95%	90%
3	6.14	6.31
6	6.06	6.24
9	5.98	6.16
12	5.90	6.08
15	5.82	6.00
18	5.74	5.92
21	5.67	5.85

Since approximately half of all lakes in the province can be classified as sensitive according to the criteria used here, 9 and 15 kg ha^{-1} yr^{-1} will protect 97.5% and 95% of all the lakes, respectively. On the other hand, 20 kg ha^{-1} yr^{-1}, the critical load originally estimated as acceptable in Ontario, will protect only 80%-85% of the sensitive lakes in the province.

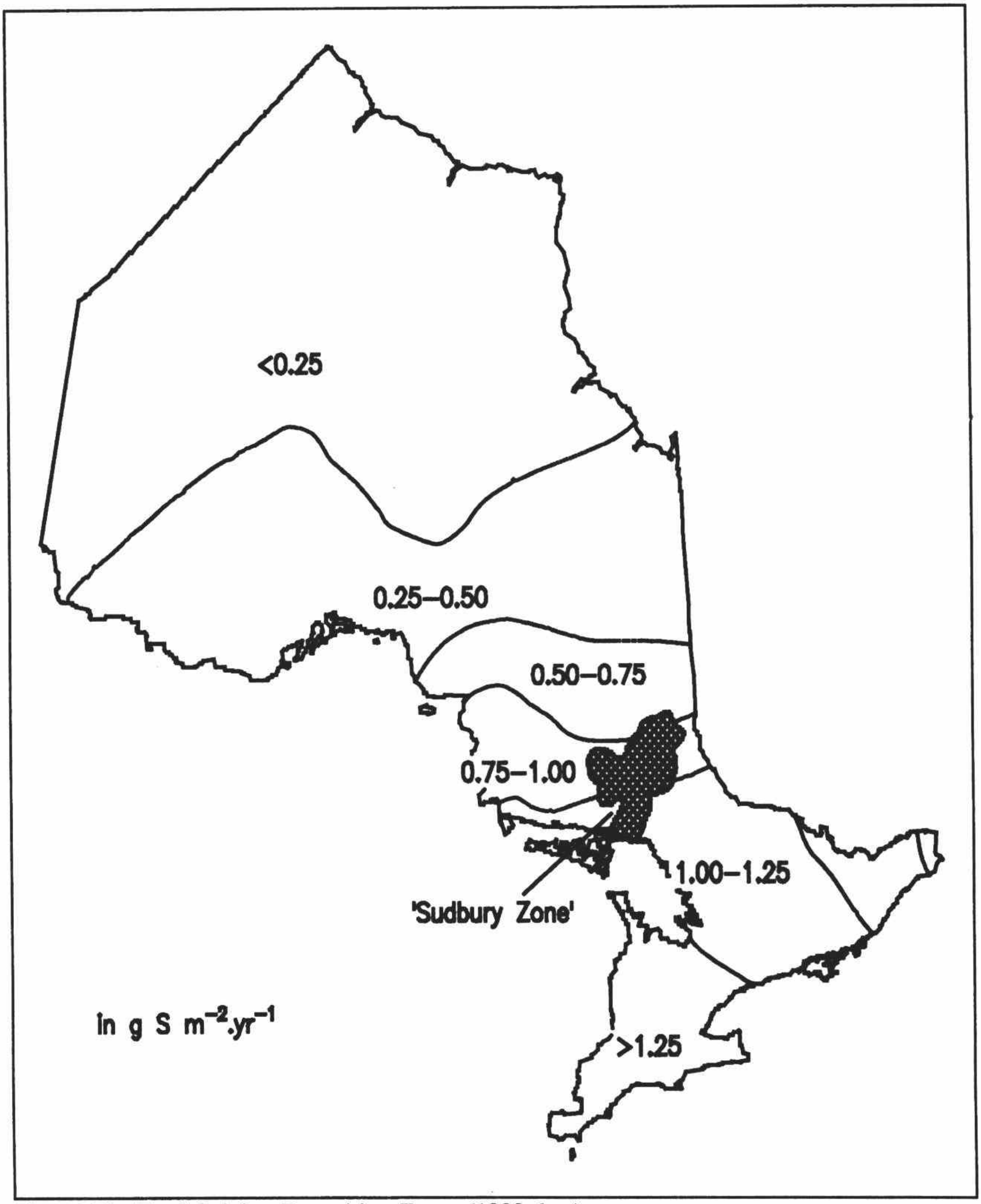

Figure 15: Total Sulphur Deposition Zones (1983 data)

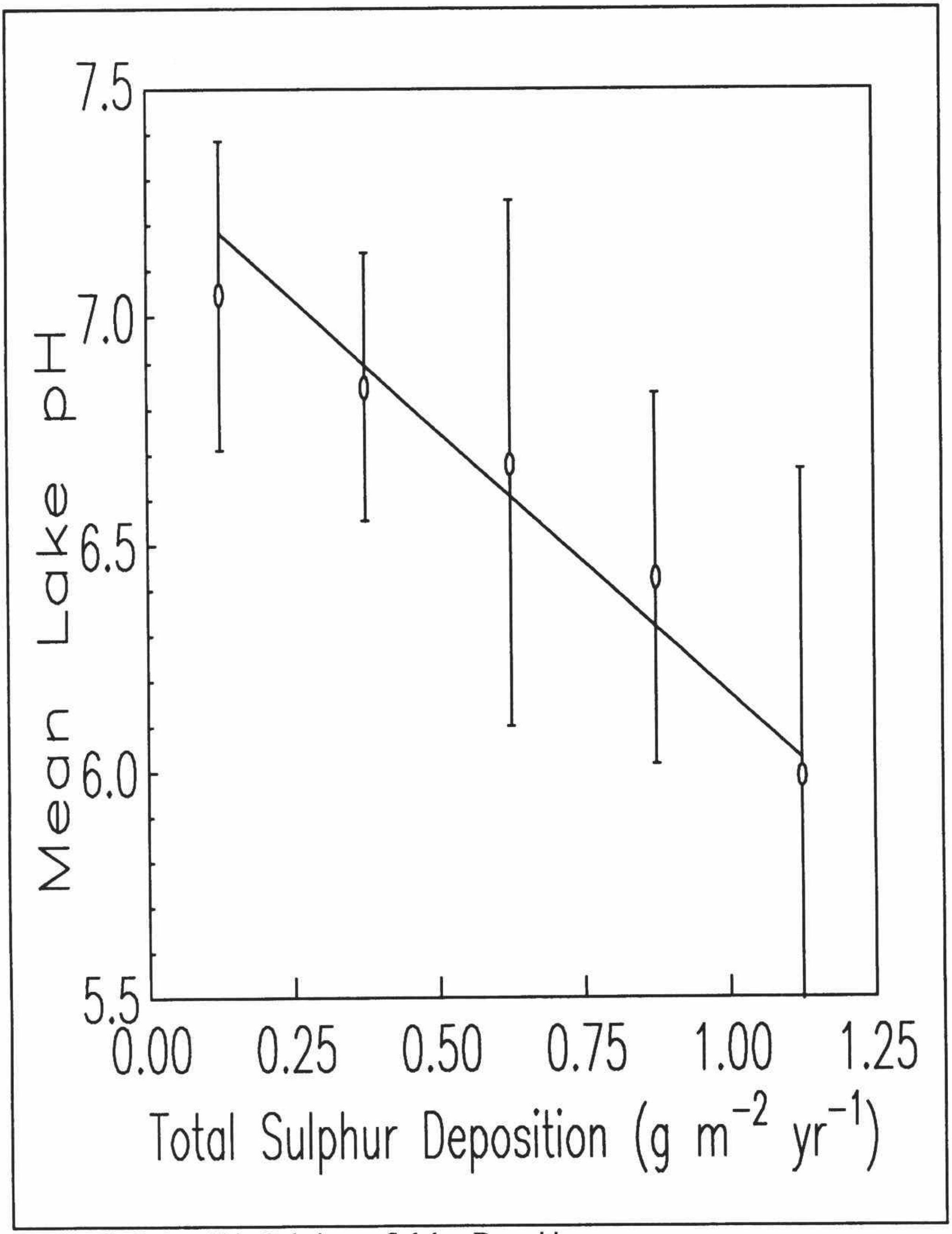

Figure 16: Lake pH in Relation to Sulphur Deposition.

B. BIOLOGICAL STUDIES

Algae

a. Paleolimnology and Lake Acidification History

In the absence of direct measurements of alkalinity loss in several Muskoka-Haliburton area lakes over several decades, indirect methods of inferring lake acidification were examined. Three algal components (chrysophyte scales, chrysophyte cysts and diatoms) all of which leave identifiable remains in lake sediments, were used to develop calibrations of algal species vs lake acid sensitivity (alkalinity, pH). In a set of 50 calibration lakes in the Muskoka-Haliburton-Parry Sound area, relationships between algal species composition in surface sediments and present day water chemistry were developed for the three groups of indicators. These were then applied to an independent set of five test lakes with present day alkalinities ranging from -0.4 mg L^{-1} yr^{-1} to 66 mg L^{-1} yr^{-1} and corresponding pH values of 4.9 and 7.9 respectively. Predictive relationships for the validation lake set were highly significant (R^2 values for predicted vs measured pH and alkalinity between 0.75 and 0.96). The best predictors were the chrysophyte cysts.

Reports were prepared in 1988 for all three calibrations (cysts, scales and diatoms). Publication of the findings is now in progress. Reports on the pH/alkalinity history of several Parry Sound and Muskoka area lakes are now in preparation. These involve application of the calibration equations to lake sediment cores. Analyses of algal indicators and ^{210}Pb dating were done at regular intervals in the core from the sediment surface to a 30 cm depth.

Considerable developmental work was necessary in order to apply these techniques. It resulted, however, in significant advancements in the taxonomy of scaled chrysophytes. This work has been recently published. The information gained has led to improved paleolimnological applications not only in this APIOS project, but also in similar studies in Europe, Scandinavia and the U.S.A.

b. Filamentous Algae

Synoptic and long term field studies of filamentous algae in acid sensitive lakes have demonstrated the propensity for certain zygnematalean species (notably *Zygogonium tunetanum* - Lievre and *Temnogametum tirupatiense* Iyengar) to proliferate as metaphytonic "clouds". A survey of 4,400 cottagers on 214 lakes in central Ontario indicated that filamentous algae are present in 48% of the lakes in the area. A statistically significant positive relationship was observed between algal presence and lake sensitivity to acidification. A high proportion (92%) of all cottagers reporting filamentous algae on their lake were concerned about reduced enjoyment of the lake.

Whole lake experiments including either neutralization (Bowland Lake) done by the Ontario Ministry of the Environment or acidification (Lake 223, Experimental Lakes Area, Kenora) done by the Federal Department of Fisheries and Oceans have demonstrated striking responses in filamentous algae.

Some additional work was done through the University of Toronto to examine the relative impacts of cyprinid, larval amphibian and crayfish grazing in lakes of different acid sensitivity. The final results of this work will be available in the near future.

Evidence from other lakes (e.g. Swan Lake, Sudbury), where acid deposition has declined in recent years as a result of decreased SO_2 emissions, suggests that filamentous algal densities have also declined.

c. Odour Causing Algae

Chrysochromulina breviturrita Nicholls was described in 1978 as a new species of planktonic prymnesiophyte occurring in Ontario softwater lakes. In 1979 and during the 1980's this species caused lake-wide "rotten-cabbage" like odours in several lakes in Ontario and four New England states. Laboratory studies of *C. breviturrita* in unialgal culture demonstrated its optimal growth requirements in low pH (5.5-6.0), low alkalinity growth media. It will not grow above pH 7. This species also has a selenium requirement and is apparently only able to utilize nitrogen in the form of NH_4^+. *C. breviturrita* has been found in about 100 Ontario lakes. Statistical analyses of a 610 lake-year data set (with and without *C. breviturrita*) showed a significant negative relationship between relative abundance of *C. breviturrita* and lake pH and a positive relationship with DOC. More than one-half of all occurrences of *C. breviturrita* were in samples from lakes with alkalinities of less than 5.8 mg $CaCO_3$ L^{-1} yr^{-1}. With only one exception, all lakes experiencing blooms and odour problems had summer pH values of 5.6 to 6.2. It is highly significant that one of these odour episodes developed in the south basin of Lake 302 near Kenora which was being acidified with sulphuric acid by the federal Department of Fisheries and Oceans. Importantly, large populations have not developed in the north basin of Lake 302 which was acidified with nitric acid (D. Findley, pers. comm.).

d. Phytoplankton Monitoring

Between 1985 and 1989 inclusive, 4,836 phytoplankton samples were analyzed from acid sensitive (and a few moderately well buffered) lakes in central, northeastern and northwestern Ontario. Most of these originated from collections by Dorset Research Centre staff in the Muskoka-Haliburton-Sudbury area. About 20% of the total sample input, however, was submitted by regional offices of Environment Ontario, the Ministry of Natural Resources' Fisheries Assessment units and the federal Department of Fisheries and Oceans.

Zooplankton

The zooplankton community of Plastic Lake was compared with that of three non-acidic reference lakes over the last decade. Inter-annual variations in community structure were larger in Plastic Lake than in the three reference lakes. The average species richness in Plastic Lake declined over the decade in comparison with the reference lakes. This indicates that the zooplankton community structure was degrading at sulphur deposition rates characteristic of the 1980's.

Daphnia galeata mendotae is one of the dominant species of crustacean zooplankton in Ontario. Abundance patterns obtained from a survey of 450 Ontario lakes were compared with thresholds of acidity determined in laboratory bioassays. This comparison demonstrated that the acidification of Ontario lakes to pH levels less than 6 has had widespread negative impacts on this important species. There are at least 20,000 such lakes in Ontario.

Other species of *Daphnia* are also sensitive to acidification. A comparison of survey and intensive lake data demonstrated that two other common species of *Daphnia* (*D. dubia* and *D. retrocurva*) are less abundant in acidified than in non-acidified lakes. Acidification impacts at pH's less than 6 can

also be seen in the ice-free season averages of species richness (the entire Crustacean zooplankton community) of 54 south-central Ontario lakes.

It has long been hypothesized that many of the effects of acidification on zooplankton have indirect causes related to altered predation pressure. Detailed examination of data from an acidic lake near Sudbury showed that intense predation from larval phantom midges can cause very unusual zooplankton community structures (i.e., almost total dominance of the community by rotifers).

Scandinavian and Ontario researchers have reported apparent differences in the impacts of acidification on larval phantom midges. A large survey of carefully selected lakes demonstrated that this apparent difference is largely attributable to the size and depth of the lakes selected. Larval phantom midges are important members of the zooplankton in small, fishless, acidic lakes in Ontario, as in Scandinavia. Indeed, phantom midges are often more important contributors to total zooplankton biomass in these lakes than microcrustacea.

It has been well established that the water quality of acidic lakes improves in response to sulphur emission reductions, but the response of zooplankton, and indeed most other aquatic biota, is unknown. A comparison of changes in zooplankton communities from 8 lakes in the Sudbury area from the mid-1970's to the mid-1980's indicated that substantial biological recovery had occurred in half of the lakes. Community structure, however, had not yet returned to the characteristics of non-acidified lakes. No recovery was recorded in some lakes that were particularly heavily contaminated with acid and metals in the mid-1970's.

The acidification of lakes influences the accumulation of cadmium by zooplankton. In clear water lakes of pH circa 5.5, that are remote from Sudbury, cadmium levels in zooplankton are elevated. This is likely due to elevated Cd levels in atmospheric deposition compared to historical levels, and because Cd export from watersheds is elevated by acidification of stream waters. Cadmium levels in zooplankton are not elevated in lakes of pH less than 5.0 despite elevated aqueous Cd levels due to competition between protons and Cd in acidic systems.

Invertebrates

Stream invertebrates (insects) have been shown to be important monitors of anthropogenic stresses such as acidification. These previous experimental studies also have indicated that short-term changes in acidity and associated aluminum concentrations could significantly alter invertebrate assemblages. During the last five years, three experimental approaches have been used to determine the influence of acidification on changes in species composition and biomass of stream invertebrates.

First, different aquatic life stages of mayflies and blackflies indigenous to the Muskoka-Haliburton districts were transplanted, prior to and during snowmelt, from streams with a range of hydrogen ion concentrations to streams with lower pH and high aluminum concentrations. Some species of mayflies were sensitive to short-term exposures to elevated acidity. Immature blackflies survived low pH (~ 4.0) and high aluminum concentrations (700 $\mu g\ L^{-1}\ yr^{-1}$) during all of the transplant experiments.

In the second experiment, additions of acid (H_2SO_4 and HCl) were added to stream reaches to simulate short-term pH

depressions in streams that have been naturally subjected to high and low deposition of acids (high=Dorset, mean H^+ deposition, 70-100 meq yr^{-1}. Low=Kenora, mean H^+ deposition of 8-10 meq yr^{-1}). Many of these invertebrates had lower survival in pH depressed streams, indicating that damage to stream invertebrates, as a result of acidification, has already occurred near Dorset, Ontario.

The final approach was to conduct surveys of insects in acid and circumneutral streams. This approach has helped corroborate the previous experimental findings. Quantitative studies of stream insect populations were made from 1937 to 1947 in Algonquin Park. Resurveys of the same locations have revealed that acid-sensitive species (mayflies and stoneflies) have decreased, and acid-tolerant species (caddisflies and blackflies) have increased in acid streams. No significant differences were found between the 1937/47 survey and the 1985 survey for non-acidic streams. The increase in blackflies, as a result of acidification has been supported by additional toxicity studies.

Toxicity Studies

The absence of a species from a lake with low pH or a population failure in an acidifying lake does not constitute proof that a species is adversely affected by acid.

Initially, toxicity testing focussed on sport fish. The lethality of H^+ and Al^{3+} to the early life stages of the following eight softwater fish species was investigated: common shiner, white sucker, smallmouth bass, lake whitefish, walleye, largemouth bass, lake trout and brook trout. Laboratory toxicity studies (as measured by reproductive success) were conducted along with the collection of population data in the field.

For all species except largemouth bass, lake trout and brook trout, the population data confirmed the laboratory studies. Common shiner were the most sensitive to acidification both in the field and laboratory, followed by white sucker and smallmouth bass. Largemouth bass, lake trout and brook trout all showed higher tolerance to acidic conditions in the laboratory than in the field. Aluminum additions in the laboratory did not significantly change the reproductive success.

Acute lethality studies of metal mixtures (Al, Cu, Zn, Fe, Mn, Ni and Pb) at ratios typical of acidified water were performed with rainbow trout and fathead minnow larvae in soft, acid water. Aluminum was more toxic to trout at pH 4.9 than any other component of the mixture, while copper was more toxic at pH 5.8. For fathead minnows, copper was responsible for toxicity at pH 5.8.

Recent data suggest that many of the small forage fish, in particular the cyprinids, are the most acid sensitive group of fish. For this reason, the sensitivity of the early life stages of cyprinids to acid in soft water was investigated.

Seven species of minnow from the Dorset area were captured and brought to the laboratory. Where possible, eggs were stripped from females of golden shiner (*Notemigonus chrysoleucas*), creek chub (*Semotilus atromaculatus*), pearl dace (*Semotilus margarita*), blacknose dace (*Rhinichthys atratulus*), and northern redbelly dace (*Chrosomus eos.*) Fathead minnow (*Pimephales promelas*) and bluntnose minnow (*P. notatus*) were cultured in the laboratory.

Eggs and larvae were exposed continually, for up to 40 days to water with a pH between 4.3 to 7.0 depending on species. With the exception of fathead minnow, mortality increased significantly and whole

body ions decreased significantly, at the lower pH's.

Individual LC_{50} varied widely across species, from about 5.9 for bluntnose minnow to less than 4.6 for pearl dace. The median LC_{50} found in the laboratory showed excellent agreement with the pH for extirpation in the field. This indicates that early life stage lethality is a good predictor of species success. Although there are clearly other determinants of cyprinid distribution in the field, these laboratory findings provide evidence of an acid effect on cyprinid distribution, and indicate that the role of other metals such as Al and Cu is probably restricted to that of a modifier.

Fathead minnow larvae were then exposed to a range of pH in static bioassays to determine the potential ameliorating effects of NaCl and $CaCl_2$. The working hypothesis was that increased Na should enhance Na influx and that increased Ca should increase gill membrane stability thereby reducing Na efflux. To overcome the difficulty of measuring unidirectional Na fluxes in organisms this small, and to determine the effect on the entire organism, survival time was used instead. In general, NaCl or $CaCl_2$ were beneficial in decreasing toxicity based on LC_{50} values.

A preliminary study was done on the effects of parasitism on the acid sensitivity in adult bluntnose minnow and northern redbelly dace. Adult fish with heavy parasite infestations showed only slight increases in acute acid sensitivity. The ET50 survival times were used since there were not enough fish for a full bioassay. Early life stages were much more sensitive to acid than adults and unless the adults show some increased sensitivity to metals, parasitism appears to play little role in determining fitness in adult cyprinids.

Biological Survey

In addition to the adverse effects of acidification on sport fish, research has also concentrated on more sensitive organisms such as benthic invertebrates. Biological surveys of softwater Shield lakes were designed to identify aquatic invertebrates that may be sensitive to low pH and to quantify their response.

The invertebrate community along the littoral zone (shoreline) was sampled on approximately 60 lakes during the fall of 1987. Water samples were also collected for chemical analysis. Additional samples were collected in the fall of 1988 to augment the data on low alkalinity lakes. Species were identified to the lowest practical taxon. The data are currently being analyzed and when completed will reveal any significant relationships between water chemistry and invertebrate distribution.

Biological Monitoring

Initial biological monitoring of lakes in the Muskoka-Haliburton region resulted in several documented cases of local, abrupt species extinctions: snails (*Amnicola*) in Heney Lake and amphipods (*Hyallela*) in Plastic Lake. In addition to H^+ sensitivity, certain life history characteristics lead to extinction over short periods of time. Extinctions may, therefore, occur when lake water chemistry degradation is slight.

Lake chemistry fluctuates on a time scale on the order of months in response to changing seasons and on the order of years in response to climatic fluctuations and atmospheric deposition. The lake biota respond differently to these changes depending on their life spans relative to the time scale of the fluctuations. Small

organisms such as algae and zooplankton, having short life spans, tend to respond mostly to seasonal changes in lake chemistry. Large organisms such as fish, with life spans of several years, may reflect changes in lake chemistry, due to acidic deposition, over a period of a number of years, perhaps decades. Benthic invertebrates, however, which have life spans of about one year respond to changes in water chemistry on an annual scale. Because these fluctuations may be large relative to the long-term changes, catastrophic collapse of such populations can occur. As a result, these organisms yield a relatively high signal:noise ratio in their response to acidic deposition.

Benthic invertebrates are easily sampled as a group with simple techniques which yield a large number of species. They are, therefore, ideal subjects for monitoring the effects of changing acidic deposition from both a practical and theoretical standpoint.

Initial work emphasized evaluation of sampling times and methods. Results of this work indicated that the fall is the best time to sample, yielding 10 times the number of organisms as the spring, using the "kick and sweep" method

Preliminary sampling of 12 lakes showed four of them to contain *Orconectes virilis*, a species of crayfish which is known to be sensitive to acid conditions. The lakes spanned a pH range from slightly acid to circumneutral and all animals caught in the most acidic lake were large adults. No young animals were caught and it appears that the crayfish in this lake are the remnants of a population on the verge of local extinction.

A federal-provincial biomonitoring program was established emphasizing benthic invertebrates in Eastern Canada. Environment Ontario contributes by monitoring 14 lakes in the Muskoka-Haliburton region of south central Ontario.

The program is too new to detect population changes, but has been successful from other standpoints. Over 500 taxa from 12 lakes were collected during the first year of sampling, and despite the use of very simple sampling techniques, the precision of relative abundance estimates is well within expected limits.

Metal and Organic Residue Monitoring

Investigations over the 1986-1990 period focused on 2 principal concerns:

a. Monitoring mercury (Hg) levels in yearling yellow perch; and

b. Investigating relationships between Hg in sportfish and environmental variables.

Some work also began in 1989 on the investigation of links between organic residues in sportfish and long range transport of organics.

a. <u>Monitoring Mercury in Yearling Yellow Perch</u>

Yearling yellow perch (*Perca flavescens*) were collected from sixteen Muskoka-Haliburton lakes to determine interrelationships between water quality, Hg residues in fish and fish condition. The lakes studied had a pH range of 5.6 to 7.3 and total inflection point alkalinities of 0.4 to 16.0 mg L^{-1}. Mercury residues in yellow perch ranged from 31 to 233 ng g^{-1} and were inversely correlated ($p < 0.001$; $r = 0.84$) with lakewater pH. Stepwise linear regression analyses selected lakewater pH as the only significant parameter associated with Hg accumulation. Alkalinity, sulphate,

calcium and dissolved organic carbon (DOC) were not significant. Likewise, lakewater pH and Hg residues in yellow perch were inversely ($p < 0.001$) correlated with fish condition. Lakewater pH accounted for 74% and Hg in fish a further 11% of the variance in fish condition. Terrestrial drainage size/lake volume ratios were also correlated ($p < 0.05$; $r = 0.78$) with Hg accumulations in perch from a subset of nine headwater lakes. No temporal trends in Hg residues were evident in yellow perch from 1978 to the present.

Fewer years of data on Hg in yearling yellow perch from lakes in the Sudbury, Chapleau and Thunder Bay areas were also collected. No temporal trends were evident in these data either.

b. Mercury in Sport Fish in Relation to Environmental Variables

Data from the Contaminants in Sport Fish Program and the Acid Sensitivity Program were used to assess the relationship between Hg residues in sport fish and environmental (primarily water quality) variables. To date, analysis has focused on lake trout, smallmouth bass, walleye and pike collected over the period 1978-1984.

Concentrations of mercury in dorsal muscle tissue of lake trout (*Salvelinus namaycush*) were positively correlated with variables indicating lake dystrophy (DOC, colour, iron, transparency) and were also correlated with watershed area and lake area. Stepwise multiple regression selected DOC as the only variable which explained a significant amount of variance (37%) in Hg concentrations in lake trout. The relationship between dissolved organic carbon and Hg appeared to be strongest in the group of lakes with values of DOC less than 4.0 mg L^{-1}. In contrast, Hg concentrations in smallmouth bass (*Micropterus dolomieui*) were correlated with variables reflecting both water hardness (magnesium, calcium, conductivity) and acidity (pH, alkalinity). The relationship was inverse for the water hardness variables and positive for acidity. Stepwise regression identified three variables significant in explaining variation in Hg in smallmouth bass: calcium, DOC and latitude.

The relationship between mercury levels in walleye (*Stizostedion vitreum*) and northern pike (*Esox lucius*) in Ontario lakes was examined. Walleye and pike occurred together in 79 of the 346 study lakes. The standard Hg concentrations in walleye and pike in these 79 overlapping lakes were highly ($p < 0.05$) correlated. The mean Hg concentration in standard length walleye and northern pike from these lakes was 0.65 µg g^{-1} and 0.52 µg g^{-1}, respectively. Lake chemical and physical parameters related to Hg concentrations in both species were examined to determine if the same environmental factors affected mercury accumulation in walleye and northern pike. Descriptors associated with lake dystrophy (DOC and iron) were positively correlated ($p < 0.05$) with Hg levels in both walleye and pike. Alkalinity, pH and hardness variables (calcium, conductivity, and magnesium) were negatively correlated ($p < 0.05$) with Hg concentrations in pike but not walleye. Latitude was positively correlated ($p < 0.05$) with Hg levels in walleye only.

Finally, the relationship between Hg in 3 species of sport fish (lake trout, smallmouth bass and walleye) and background levels of Hg in lake sediment was examined. Using background sediment Hg as a reflection of geological Hg levels, the analyses suggested that lake to lake differences in Hg levels in fish cannot be accounted for by differences in geological Hg.

Taken together, the data support the premise

characteristics act together to influence Hg levels in fish. Common elements do emerge. Lakes with higher DOC levels tend to have sport fish with higher Hg levels. Lakes with lower pH and alkalinity tend to have sport fish with higher Hg levels. Lakes of lower hardness (lower Ca, Mg, Conductivity) tend to have sport fish with higher Hg levels.

c. Organic Residues in Fish: Relationships to Long Range Transport

Work began in FY 1989/90 on the subject of the long range transport of organics and the significance of this pathway to organic residue accumulations in sport fish. In a preliminary assessment, PCB and total DDT concentrations in sport fish were found to be highly correlated for most lakes, suggesting a common source for these contaminants. A lack of correlation was useful in identifying lakes where local point source PCB contamination had occurred. The presence of planktivorous fish was associated with elevated PCB and DDT concentrations in lake trout.

MNR Fisheries Acidification Program

a. Extensive Surveys

This program has used extensive survey data to examine relationships between fish species presence/absence and lake physical/chemical parameters. Fish survey data were gathered on 9000 lakes and 3200 of these lakes had recent chemical data. The total number of species was found to increase with lake area, whereas the number of cyprinids (minnow species) was generally unrelated to lake size. When the effects of lake size were taken into account, a decline in number of fish species occurred below pH 6.0. Cyprinids appear to be very sensitive to low pH as their occurrence declined below pH 6.0. The distribution of each species was examined to evaluate the potential of the 13 most common small (≤ 10 cm total length) fish species to be used as early indicators of change in the fish community due to acidification. The most promising species were common shiner (*Notropis cornutus*), fathead minnow (*Pimephales promelas*), bluntnose minnow (*Pimephales notatus*), blacknose shiner (*Notropis heterolepis*) and slimy sculpin (*Cottus cognatus*) (Figure 17).

Losses of sport fish populations in Ontario were calculated using, 1) observed pH thresholds for mortality based on survey data (pH <5.5 for lake trout, walleye and smallmouth bass; pH <5.0 for brook trout, *Salvelinus fontinalis*), 2) atlases describing species' distributions, and 3) the extensive Ontario chemical data set. The estimated number of lakes that have either lost their former populations or have had residual non-reproducing populations in Ontario were 119 lake trout, 39 brook trout, 52 smallmouth bass, and 14 walleye lakes (Table 2).

Table 2
Estimated Number of Sport Fish Lakes in Ontario Affected by Acidification

	SPECIES			
Number of Lakes	LT	BT	SMB	W
Total # in Ontario	2,318	2,100	2,421	4,038
# Sampled for pH	1,296	639	901	989
# Sampled with pH < species thresholds	78	8	27	7
Estimated # with pH < species thresholds	119	39	52	14
# Affected lakes in Sudbury Area	94	14	18	8

LT = Lake Trout BT = Brook Trout
SMB = Smallmouth Bass W = Walleye

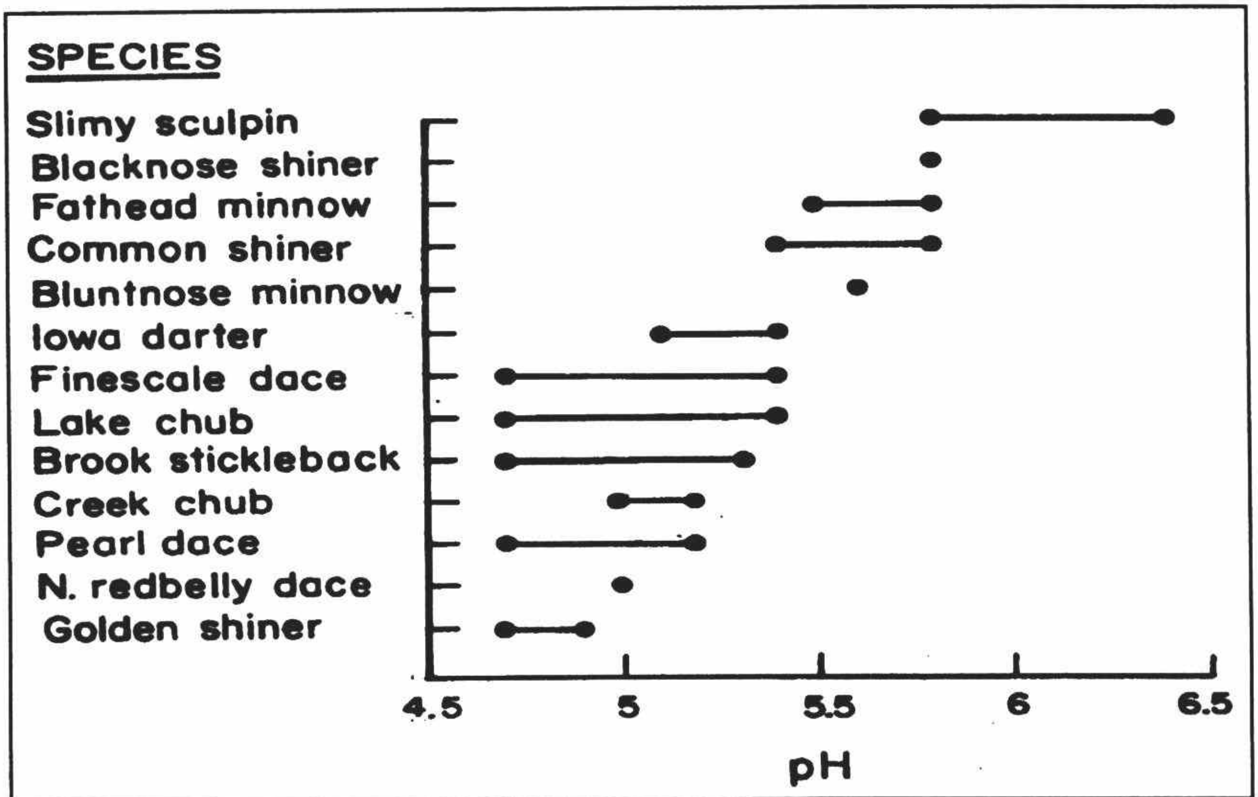

Figure 17 Change in Fish Communities Due to Acidification

b. <u>Comparative Lakes</u>

Netting surveys and field bioassays were conducted in many Northeastern Region lakes to generate the pH thresholds for survival and reproduction of lake trout, brook trout, smallmouth bass, and walleye. These thresholds were used to derive the provincial estimates of sport fish resources affected by acid rain.

Recently, many of these lakes have been resurveyed to examine the recovery rates of affected fish populations. Many of these lakes are affected by sulphur dioxide emissions from the Sudbury smelters. Emission reductions have led to improvements in water quality in many Northeastern Region lakes. In Whitepine Lake, where a residual population of non-reproducing adult lake trout occurred, recruitment resumed when lake pH rose above pH 5.5. Increased competition and predation by lake trout then resulted in the collapse of a large population of yellow perch. In Joe Lake, a reproducing brook trout population was re-established through stocking after water quality improved. Lake pH has improved in 25 former lake trout lakes in the Sudbury area, allowing fishery managers to stock hatchery reared lake trout and begin rehabilitation of sport fisheries in this area.

c. <u>Trend-Through-Time-Studies</u>

Field studies located in the Muskoka/Haliburton area have assessed the status of fish populations in eighteen (18) low alkalinity lakes. A subset of these lakes will be used to assess the impact of future acid deposition levels on fish populations in this acid sensitive area of Ontario.

These studies have found that acid sensitive species such as common shiner, fathead

minnow, bluntnose minnow and blacknose shiner, are present in lakes with pH >5.5. Acid tolerant species such as yellow perch, northern redbelly dace (*Chrosomus eos*), golden shiner (*Notemigonus chrysoleucas*), and pearl dace (*Semotilus margarita*), are present in low pH lakes (lakes with pH <5.5 and >5.3).

MNR Wildlife Studies

The objectives of this study were to:

a. Determine the association of wetland acidity with reproductive parameters and insect prey of the Eastern Kingbird near Sudbury, Ontario.

b. Determine the contribution of inorganic and organic acids to the acidity of wetlands in the Killarney area.

c. Determine the toxicity of aluminum to amphibians in waters containing natural organic acids.

d. Determine the levels of cadmium in the common goldeneye.

e. Assess the influence of lake acidification on the reproductive success of the common loon in Ontario.

a. <u>Wetland Acidity and the Eastern Kingbird</u>

A three year study investigated the relationship between water quality variables associated with acidification and the reproduction of the Eastern Kingbird near Sudbury. The major percentage of variation in Kingbird reproductive factors measured (including egg weight loss and bone growth), was explained by genetic differences between siblings or by environmental differences between nests. However, an additional amount of variation was explained by lake acidification. Metal enrichment of aquatic prey insects was related to acidity in the study area. However, no food limitation for the Kingbird could be attributed to lake acidity.

b. <u>Sources of Acidity in Killarney Wetlands</u>

The role of natural or organic acidity in Killarney area wetlands was addressed in another study which was published in 1986. Data indicate that acidity comes predominately from sulphates and that organic acidity plays a minor role in the Killarney study sites.

c. <u>Aluminum Toxicity to Amphibians</u>

Further surveys of amphibian habitat in the Sudbury area were carried out in 1986 and 1987 to determine if organic compounds in acidic pond water ameliorate the toxic effects of aluminum and other metals on amphibians. Laboratory and field survey information suggests that aluminum and copper are not toxic to amphibians in ponds containing even low levels of dissolved organic compounds while organic compounds are probably toxic to amphibians in low pH, dark water ponds.

d. <u>Cadmium Levels in the Common Goldeneye</u>

A program to determine the levels of cadmium present in the common goldeneye (*Bucephala clangula*) and to test the feasibility of using feathers as a reliable, non-fatal field sampling technique to monitor cadmium burden was carried out with Trent University from 1986-1988. Results indicated that feather cadmium levels of Ontario goldeneye were higher than those reported elsewhere. Cadmium levels were greater in females than males, but no difference was measured between juveniles

and adults.

e. Lake Acidification and the Common Loon

A joint program with the Long Point Bird Observatory from 1989 to 1992 is to examine the mechanisms responsible for the effects of acidity on the reproductive success of the Common Loon (*Gavia immer*). The use of observations of feeding behaviour and reproductive performance of this species as an indicator of water quality in lakes will also be assessed. Preliminary results show evidence of lower loon chick production (fewer lakes producing six week old chicks) in more acid sensitive lakes. There may be an optimum alkalinity (about 5 or 6 mg L^{-1}) at which adult loons spend the least effort feeding their chicks. More efforts were spent feeding chicks in lakes with higher aluminum concentrations.

C. REMEDIAL METHODOLOGIES DEVELOPMENT

Two lakes in central Ontario, Bowland and Trout Lakes, were limed to examine the feasibility of using liming as a mitigative measure for lakes acidified by anthropogenic acidification.

Bowland Lake, 70 km north of Sudbury, was a clear, oligotrophic lake with a flushing rate of ~ 2 years, which had once supported a native lake trout population. In 1982 it had pH <5.0, total Al of 130-150 µg L^{-1} and a population of yellow perch but no lake trout was found in index netting. A trial introduction of lake trout before liming had failed. Bowland Lake was limed in August 1983 using 84 tonnes of calcite dropped as a dry powder from a Canso aircraft.

Trout Lake, 50 km north of Parry Sound, was clear, marginally acidic (pH 5.8-6.2) with low total Al (25-40 µg L^{-1}). In 1982 it supported a complex community of 16 species of fishes including lake trout and smallmouth bass; however, the pH had reached 5.8 to 6.0 and severe spring melt episodes of very acidic melt water (pH <4.5) were occurring. Trout Lake was also limed in May 1984 with 135 tonnes of slurried calcite dropped from a Canso.

The Acidic Lake: Bowland Lake

After the calcite addition, the pH in Bowland Lake increased from <5.0 to 6.7, alkalinity increased from -0.3 to 4.5 mg L^{-1} and Al declined gradually to 30 mg L^{-1} (Figure 18a, 18b). By 1989, six years after liming, lake flushing had reduced the pH to 5.7 and alkalinity to 1.5 mg L^{-1}, Al had risen to 60 mg L^{-1}, DOC remained higher than pre-liming and Secchi depth continued to be less than pre-liming.

The biological community of Bowland Lake changed after liming. Lake trout were successfully introduced as adults and fingerlings the fall after liming. Bioassays of caged lake trout fry and fingerlings which had shown <10% survival before liming resulted in 100% survival after liming. Lake trout eggs had also shown <10% survival in the acidic lake in overwinter bioassays. Survival of eggs increased to >75% in the winter and spring after liming and stayed over 50% for the next six years of sampling - comparable to the results on a circumneutral reference lake. Successful recruitment by the stocked lake trout was documented when three and four year old fish were sampled which could only have been from natural recruitment.

Growth of stocked lake trout fingerlings in limed Bowland Lake was comparable to other lakes in the region, however, adult

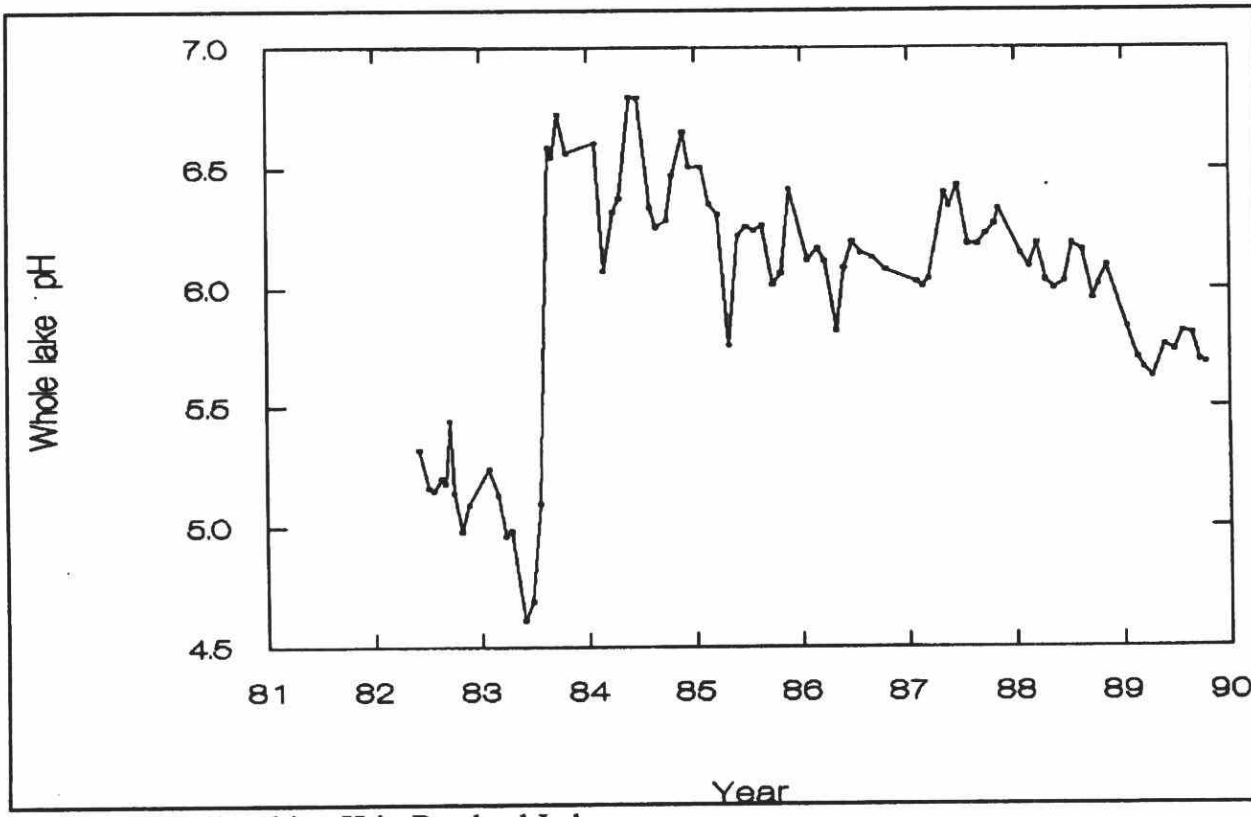

Figure 18a Trend in pH in Bowland Lake

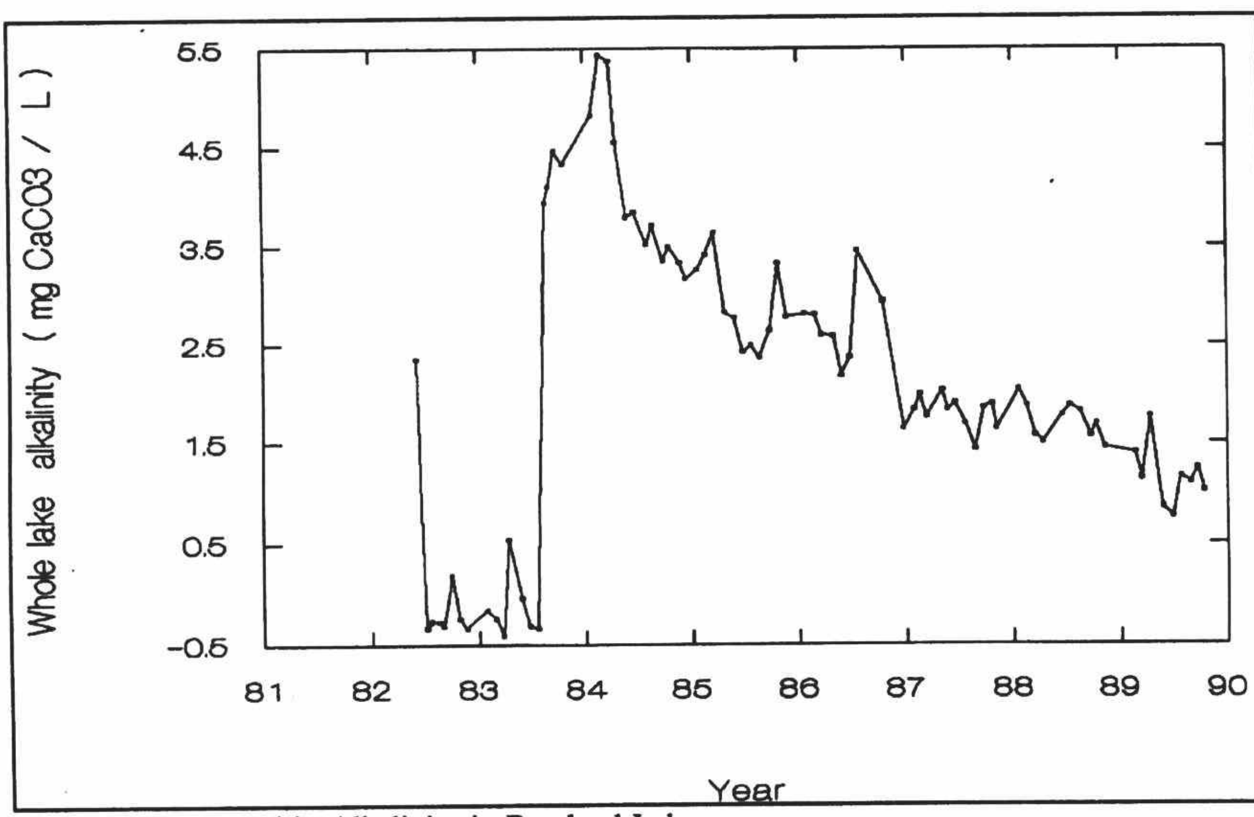

Figure 18b Trend in Alkalinity in Bowland Lake

trout, which had been transferred from circumneutral lakes, lost condition and died over a six year period perhaps indicating their inability to adapt to the Bowland Lake forage base. The yellow perch population appeared to be driven primarily by lake summer temperatures and lake trout predation. A strong 1983 year class resulting from high summer temperatures, drove the population of >1+ perch to a high of 353,000 in 1985. Gradually, natural mortality and predation reduced the population to 76,000 by 1989. Preliminary results from application of a fish bioenergetics model suggest that year class strength of yellow perch is a driving force in the lake's ecology. Small yellow perch feed on zooplankton and zoobenthos as well as being a source of food for larger perch and lake trout. Annual variation in this population will effect the direction and efficiency of energy flow through the system.

Phytoplankton biovolume in the photic zone did not change with liming. Analysis using multivariate ordination techniques is under way to identify possible compositional changes that may have occurred in the plankton. Zooplankton composition was affected by liming. Certain acidophilic species decreased in abundance and certain species preferring less acidic conditions either became more abundant or appeared. The abundance of rotifers and ciliates generally increased after liming whereas the abundance of crustaceans declined slightly.

Extensive sampling of the zoobenthos occurred in the four years after liming. Five sampling strategies were applied in late summer to sample a range of habitats to determine the species richness and abundance of benthic invertebrates. The species richness of benthic invertebrates was consistently higher in the post liming years. There was a notable failure of certain important types of acid sensitive invertebrates including gastropods and amphipods, to recolonize Bowland Lake up to 5 years after liming. The abundance and biomass of invertebrates declined in hypolimnetic sediments. The size specific decline in chironomids in hypolimnetic sediments, responsible for the overall decline in community biomass, is thought to be a consequence of lake trout feeding which began concurrent with lake neutralization.

The Endangered Lake: Trout Lake

The pH and alkalinity in Trout Lake increased to 6.6 and 2.6 mg L^{-1}, respectively, immediately after liming then were reduced by lake flushing to 6.0 and 2.0 after six years (Figure 19a, 19b). Total Al levels were reduced by 40% to ~25 mg L^{-1}. Ca concentrations rose from 2.2 mg L^{-1} to 3.8 mg L^{-1} directly after liming then dropped to 2.6 mg L^{-1} by 1989. DOC, Secchi depth and manganese remained unchanged by liming.

The biological community did not show as great a change as in the acidic lake however some important changes did occur. In the phytoplankton community, while the peak summer biovolume did not change, ordination analysis clearly segregated three groups of samples: a preliming group (1983/4), an intermediate early postliming group (1985/6) and a late postliming group (1987-89). While phytoplankton biovolume was predominantly diatoms throughout the study, the biovolume of chrysophytes declined after liming and the cryptophytes increased. Zooplankton abundance and biomass declined in the first three summers after liming then returned to preliming levels. Rotifers became more abundant in the plankton starting three years after liming.

Average ice free abundances of crustaceans were reduced the first three summers and then abundances returned to preliming levels.

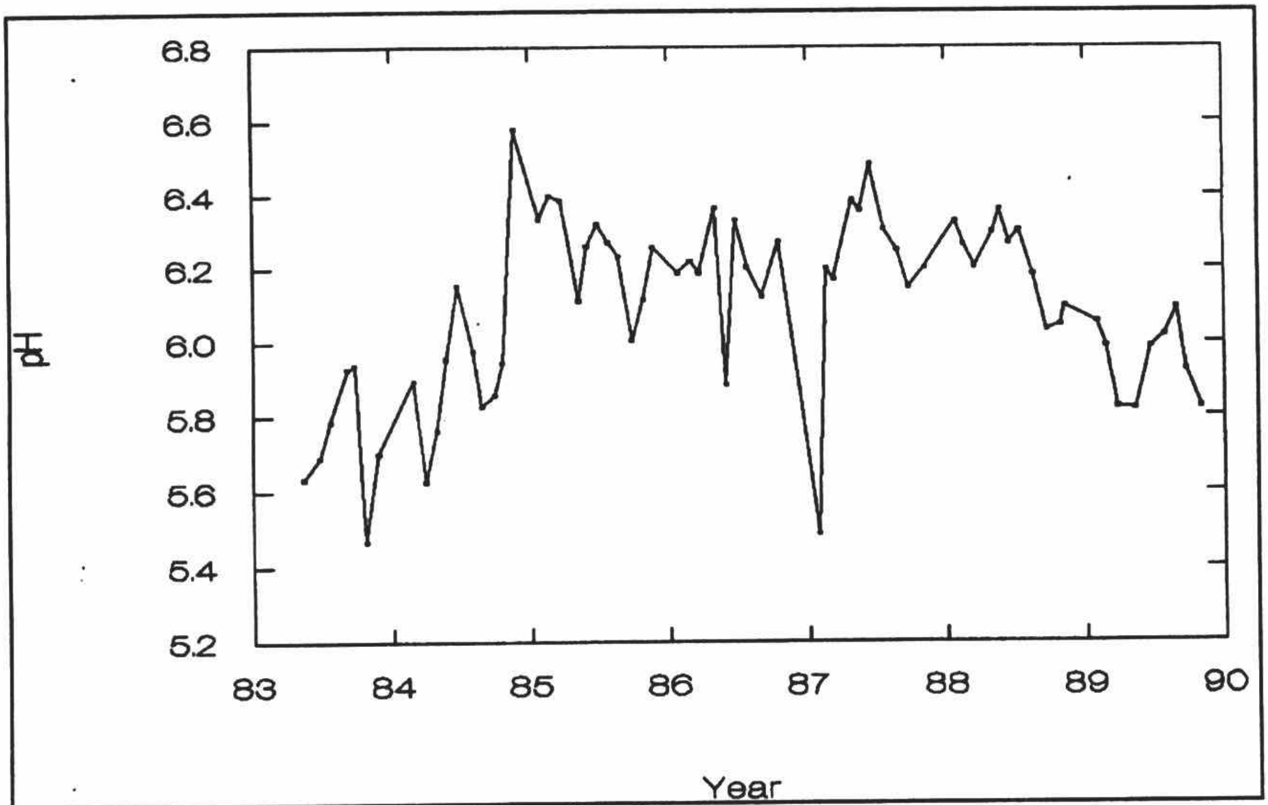

Figure 19a Trend in pH in Trout Lake

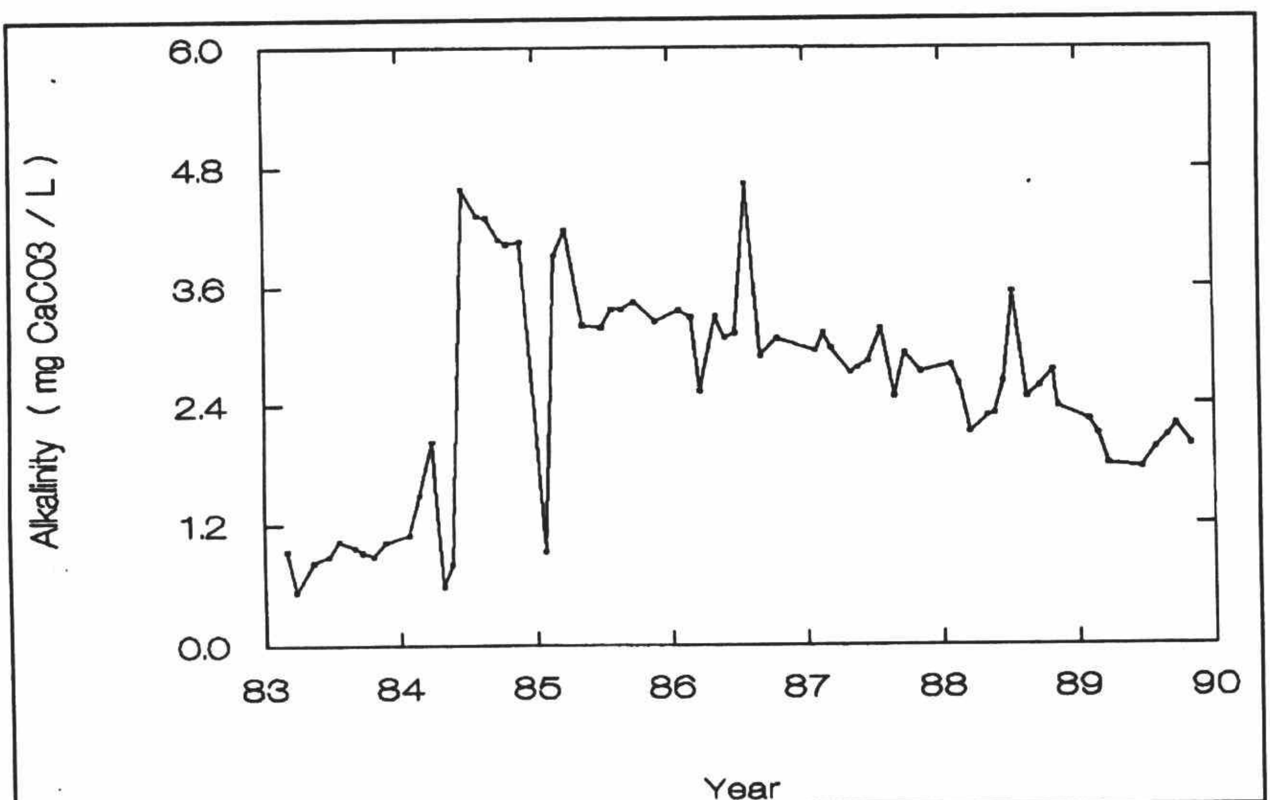

Figure 19b Trend in Alkalinity in Trout Lake

Ciliate abundance and biomass did not appear to be affected by liming. Species composition changes are being investigated. The population of *Mysis relicta* increased two-fold after liming and remained high throughout the study period. This may have had important implications for the lake trout, for which this is a favourite prey. Lake trout growth rates increased significantly in Trout Lake beginning the year after liming. Other nearby lake trout populations did not exhibit increased growth rates (Figure 20). Fluctuations in the abundance and growth rates of other species of fishes in Trout Lake did not show consistent trends.

Summary & Conclusions

Liming was successful in increasing the pH and alkalinity of both lakes for a limited period, determined primarily by the lake flushing rate. Aluminum was reduced from potentially toxic levels in the more acidic lake and while levels increased slowly, they did not return to preliming levels at the same rate as the pH and alkalinity. The improved water quality in both lakes had effects on the biota. In the acid lake these effects were most dramatic as lake trout could be reintroduced into the system. Many of the other changes seen in the ecological community in previously acidic Bowland Lake were in response to the addition of lake trout rather than directly attributable to improved water chemistry. In Trout Lake, where there were no documented signs of stress on the lakes ecosystem prior to liming, shifts occurred in the phytoplankton and zooplankton communities species composition and in the abundance of *Mysis*. The increase in *Mysis* may have been responsible for an increase in the growth rate of lake trout after liming. While successful recruitment of lake trout in Bowland Lake has been documented, no information is yet available regarding the rates of success of indigenous lake trout reproduction in either lake.

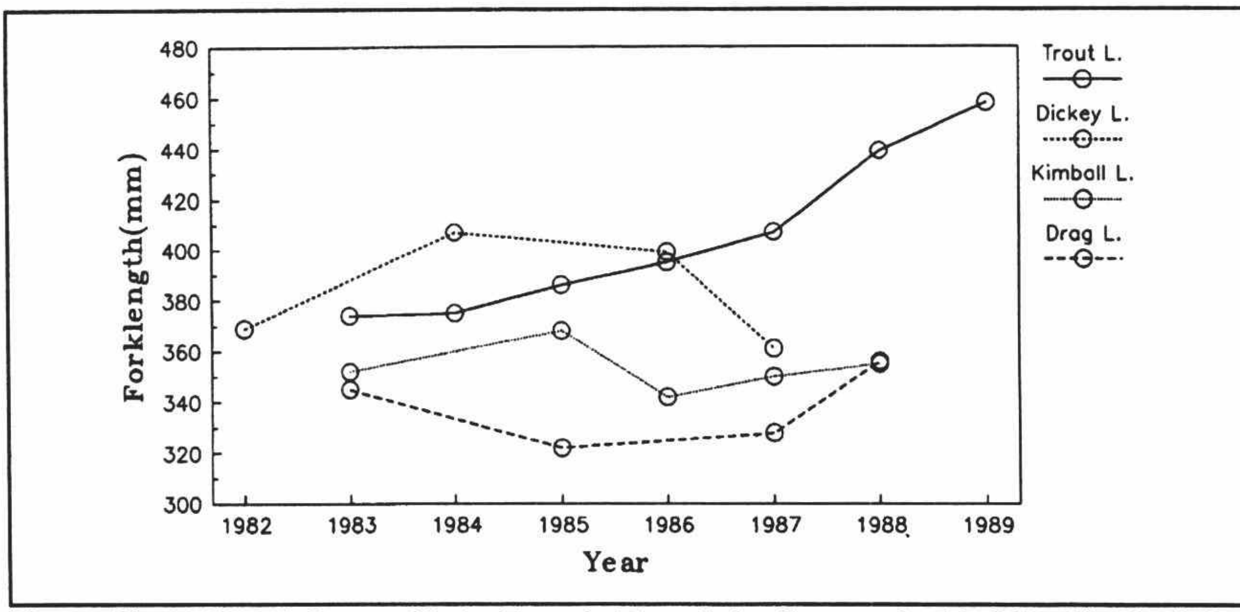

Figure 20 Growth Rates of Lake Trout in Trout Lake Compared with Other Nearby Lakes

Terrestrial Effects Studies (Task 3)

Contact: W.D. McIlveen

The primary objective of the Terrestrial Effects program is to determine the impact (if any) of acidic precipitation and related pollutants on the terrestrial environment. The program is divided into several sub tasks: Vegetation; Soils; Forest Productivity and Decline (responsibility of the Ontario Ministry of the Environment); and Wildlife (responsibility of the Ontario Ministry of Natural Resources).

A. VEGETATION STUDIES

Three primary objectives were identified:

1. the establishment of baseline chemistry of tree foliage at a number of sites across Ontario,

2. the performance of controlled environment exposures of agricultural crops and forest species to acidic precipitation, and

3. the establishment of a network for monitoring changes in lichen and moss flora that might result from acidic precipitation and related pollutants.

Tree Foliage Chemistry

In Southern Ontario, tree foliage chemistry was established at twelve sites in 1980-81. The same trees were sampled again in 1986. A set of 12 tree species at 14 locations within the High Falls biogeochemical study (Northeast Region) area were sampled. Foliage samples were collected at 50 sites across Northwestern Ontario. The data form a basis for future reassessment to determine nutritional and toxic stresses associated with acidic precipitation.

Agricultural crops

In 1984, a greenhouse experiment was conducted to determine the effects of simulated acid rain (SAR) on alfalfa, barley, cabbage, corn, cucumber, and soybean. The crops were exposed to simulated acid rain as low as pH 2.6 and as high as pH 5.6. Little effect was found on the dry biomass accumulation of shoot and root components with the exception of cabbage. The results demonstrated that plant response to SAR is not only species dependent, but also cultivar dependent. The results of a collaborative greenhouse study designed to determine the effects of SAR on Cherry Belle radish plants indicated that there was a 10 percent reduction in mass at pH 3.3 for hypocotyls but there was no significant effect on shoots.

A field study was conducted in 1985 and 1986 to investigate the effect of SAR on yield of soybean (*Glycine max* cv. Hodgson), using the Rain Exclusion Canopy (REC) System at the Phytotoxicology Laboratory in Brampton, Ontario. Five concentrations of SAR were used ranging in pH from 3.0 to 4.3. Results of the 1985 study showed a slight increase in yield in the more acidic treatments. However, the 1986 study results

did not show significant treatment effects on seed yield or other components of yield. Yield was greater in 1985 than in 1986, and was attributable to moisture conditions and not to SAR treatments.

Forest Tree Seedlings

The focus of study shifted from agricultural crops to forest seedlings in 1987. Two tree species were used, sugar maple (*Acer saccharum*) and white spruce (*Picea glauca*). Sugar maples were chosen since there was concern in Ontario that this species might suffer decline similar to that in Quebec. White spruce is a commercially important species in Ontario and research in New Brunswick had indicated that it might be as sensitive to acid rain as red spruce which was showing severe decline in the Appalachian mountains in the United States.

A preliminary greenhouse experiment was conducted in 1987 exposing sugar maple and white spruce seedlings to SAR of pH 3.3 or 4.3 (2.4 cm per week) and to ozone of either 0 or 100 ppb (seven hours exposure twice a week). The treatments had no effect on overall height, apical growth, side-branch growth, number of leaves, leaf area or stem diameter of sugar maple seedlings. Maple foliar calcium concentration was highest in leaves exposed to SAR alone and lowest in seedlings exposed to both simulated acid rain and ozone. The pH 4.3-sprayed seedlings had consistently higher foliar calcium concentration compared to those sprayed with SAR alone. Maple seedlings exposed to ozone alone or pH 3.4 and ozone had significantly lower stomatal conductance than the plants sprayed with pH 4.3 SAR. Spruce seedlings showed no significant effects due to the treatments.

The REC system was used to study the effect of acidic deposition on sugar maple and white spruce seedlings. The trees were subjected to three different SAR treatments (pH 3.2, 4.3 or 5.6) at regular intervals. Non-destructive measurements on the seedlings included: 1) visible injury rating; 2) photosynthesis rates; 3) throughfall and soil leachate chemistry; and 4) plant height, stem diameter, leaf number. Destructive sampling of selected seedlings was made in 1989 to investigate: 1) pigment content of leaf tissues; and 2) physical and chemical changes in roots and foliage. The results indicate no significant effects due to simulated acid rain in either species or in any of the soils. There was a trend of decreased growth and photosynthesis in the pH 3.2 treated plants compared to the pH 4.3 or 5.6 treatments.

The rain exclusion canopy system was designed to apply both SAR and ozone. Owing to technical problems, adequate filtration for clean air treatments could not be attained, therefore, interpretation of the experimental results including ozone as a factor would be misleading.

Acidic Rain - Insect Defoliation Study

Much of the severe dieback of sugar maple which occurred in Ontario was associated with defoliation by forest tent caterpillar. It is possible that stressed trees may be preferred hosts of insects. The growth rates of the insects on foliage treated with SAR at pH 3.2 was greater than on controls treated at pH 5.6 and the insects matured earlier. Growth and survival of trees was reduced following insect defoliation.

Lichen and Moss Study

The concentrations of 20 elements were measured in the lichens *Cladina mitis* and *C. rangiferina* and in the moss *Pleurozium schreberi* at 44 locations throughout Ontario. The spatial patterns of elemental concentrations suggest that they were

partially related to acidic precipitation chemistry and partially to local sources of pollution. Regional gradients of Ni, Pb and sulphur concentrations in *Cladina* showed the highest concentrations in central and north-central Ontario and appear to reflect the chemistry of precipitation. Lichen flora examined at four biogeochemistry study sites indicated that compositional differences were present. These differences might be attributable to historic and more recent sulphur deposition.

B. SOIL STUDIES

In the period 1985 - 1990, the soils studies consisted of four main activities. The primary objective of the soil studies was to determine if acidic precipitation was altering soil chemistry and ultimately plant growth.

Soil Baseline Study

In 1980 - 1981, 250 soil profiles were examined across Ontario and chemical and physical parameters determined for each horizon. This data base provides a baseline for future monitoring. Resampling of 100 of these sites was conducted in 1987. This generated over 700 samples which were then analyzed for the same parameters determined in 1980 - 1981. Preliminary statistical analysis of the data indicates that some changes may be occurring over time in some soil properties. For example, the pH has decreased on the majority of soils. Since the implications of a change in pH over such a short period of time are of major concern, some original samples from 1980 and from 1987 are being reanalysed simultaneously to assure that the trend is not due to changes in analytical test methods.

To assist in the interpretation of the soil baseline information and in the planning of future soil monitoring studies, a soil variability study was undertaken in 1982 - 1983. The results of the statistical analyses, completed in 1989, give some useful estimates of the degree of soil variability that can be expected on some Shield soils in Ontario which are sensitive to acidic deposition. However, analysis of changes in soil properties over time, that is, temporal variability, indicated that the sampling methodology may have in itself been influencing some chemical and physical parameters. It appears that observations made may not have been independent of the procedure of observing. For future attempts at observing soil variability in time, it must be suggested that the experimenter rely on the random nature of soil characteristics over an area and have many sites appropriately located such that one observation can not influence the next observation in time.

Soil Sensitivity

In collaboration with the Lands Directorate of Environment Canada a 1:1,000,000 scale colour map of Ontario was produced in 1986 illustrating relative ecosystem sensitivity to acidic deposition. The map shows areas interpreted, according to soil depth, soil chemistry, and bedrock characteristics, to have high, moderate or low potential to reduce the acidity of atmospheric depositions. It can be used to interpret regional surface water sensitivity to acidic deposition. This compilation shows Ontario to consist of the land classes shown in Table 3.

Table 3
Summary of Classification of Terrestrial Sensitivity to Acidic Deposition in Ontario

Class	Total Area (km^2)	% of Province
Non-sensitive	247,000	23.1
Moderately Sensitive	192,000	18.0
Highly Sensitive	335,000	31.3
Organic (not rated)	295,000	27.6

The document also discusses specific, potential terrestrial sensitivities and implications for forest productivity.

Open Top Soil Columns

In 1983, differing soils from three locations across Ontario were collected and placed into soil columns. These columns were distributed to three locations with contrasting acid deposition loads such that each location had soils of the three different soil types. These columns were placed in the ground with the open tops exposed to the natural elements including acidic precipitation. Sufficient numbers of columns were established to allow for the monitoring of chemistry changes in two columns every five years for 20 years. The first of these columns was collected and sampled in 1988 and data analysis is underway.

Sugar Maple Nutrition Studies

One theory explaining the observed forest declines in Ontario and Quebec is that accelerated leaching of nutrients from forest soils, caused by acidic deposition, has resulted in tree nutrient deficiencies and consequent decline symptoms. To determine whether the condition of maple forests can be improved through enhancement of the soil nutrient status, an experiment was set up in 1988 in cooperation with the Ontario Maple Syrup Producers Association. Seven different fertilizer treatments were applied to healthy and declining trees at 4 different sites. Prior to fertilization treatment, all trees were assessed for decline status and sampled for chemical analysis. The soil under each tree was analyzed for nutritional status. Decline assessment sample collections and chemical analyses were performed again in 1989, the year following the application (in fall) of the fertilizer treatments. The condition of the experimental trees will be assessed periodically.

C. FOREST PRODUCTIVITY AND DECLINE STUDIES

The objectives of the Forest Productivity and Decline Studies included the determination of the role of acidic precipitation in the decline of sugar maple trees in the Muskoka-Parry Sound Districts of Ontario and the relationship between acidic deposition and forest productivity.

Hardwood Observation Plots:

In FY 1985/86 and FY 1986/87, a study was initiated to determine the incidence and severity of hardwood decline across the province of Ontario. One hundred and ten

permanent observation plots dominated by sugar maple and consisting of 100 trees each were established across the province. Each tree within the plot was assessed according to the number of dead branches, chlorosis and undersized leaves in the crown. The data were used to calculate a Decline Index using 0 to represent a tree without any dieback and 100 to represent a dead tree. Data from the original evaluation indicate the presence of plots with moderate to severe dieback in the southwestern part of the province as well as the Niagara and Muskoka areas. Dieback is scattered through the northern part of the study area and there is a broad band of relatively-healthy hardwood stands extending from Lake Huron south of Georgian Bay east to the Ottawa area. The pattern of dieback could not be directly related to acidic precipitation patterns; however, soil characteristics appear to play an important role in determining patterns. The severity and extent of dieback was greatest where soil had little buffering capacity and acidic deposition was highest. The data also indicate that several hardwood species are experiencing a greater degree of decline than maple.

In 1987 and 1989, the plots were reexamined to determine whether the condition of the trees as measured by the Decline Index had changed. The 1987 survey identified some plots where the Decline Index was higher than in 1986. These areas correspond to known or suspected forest insect infestations. For the majority of the plots not affected by defoliating insects, the mean Decline Index of trees did not change by more than one standard deviation between 1986 and 1987. Between 1987 and 1989, 16% of all survey plots showed increased decline while over half of the plots (55%) showed an improvement in health. There was no evidence of a direct relationship between tree decline and wet sulphate deposition patterns.

Maple Decline Etiology

A study was initiated in 1984 and continued through 1989 to determine the role of acidic deposition in the decline of woodlots managed for maple syrup production in the Muskoka/Haliburton area of Ontario. Eleven permanent observation plots were established to monitor tree condition. In general, the condition of the trees remained the same or improved slightly from 1984 to 1987. In 1988, however, the degree of decline in Southern Ontario areas was the highest of any year measured. The primary reason for the higher decline index was an increase in the degree of yellowing of foliage, partially attributable to the drought conditions which prevailed in 1988 and, in some cases, to the effects of insect defoliation. A slight improvement, relative to 1988 only, was noted in 1989 in Muskoka while deterioration continued to increase at the Peterborough sites.

Determining the cause of tree dieback at each site is difficult owing to the many stresses that are or were in effect prior to the dieback. Severe defoliation by forest tent caterpillar in the late 1970's was combined with spring droughts in 1976, 1977 and 1983. Armillaria root rot, tree age, site management and possibly the additional stress of acidic deposition, may be responsible for the observed poor tree condition.

Hardwood Growth Studies

The lumber industry is of prime importance in Ontario; therefore, anything that reduces the rate of growth of trees is a major concern. Our studies, focusing on the growth of sugar maple, included detailed stem analysis and increment core measurements of outwardly-healthy hardwood trees at several locations. These included 42 plots in six general areas across

a pollution gradient in the Province of Ontario. Supplemental data were obtained at several other study plot locations.

Growth rates of sugar maple and other species were observed to decrease sharply in about 1960 (Figure 21). The greatest reductions in relative growth (comparison of growth from 1900 to 1939 with growth from 1955 to 1985) were observed in zones with the highest levels of acidic deposition and ozone concentrations. Specific volume increments were reduced by approximately half between 1960 and 1986 in maple trees using stem analysis techniques. A comparison of the ring widths for over 1000 trees over the 30 year period starting in 1901 (when pollution was low) with a more recent 30-year period ending in 1984, shows that growth in the zone of highest deposition has decreased by 27%. Growth has increased in the zone of low deposition.

The analysis of models of sugar maple growth and climate revealed a number of interesting patterns. Across all time periods (1900-1989), climate was found to best predict growth in the Barrie area followed by Peterborough and St. Williams. The reverse pattern occurs for the percent contribution of previous years growth for the same period. These results suggest that environmental factors other than climate have had an increasing importance in predicting growth across the study areas. This may in part predict a parallel gradient in the suitability of sites (soil depth, soil chemistry, etc) to support maple growth. In addition, this trend may reflect the location of the study sites in major pollution zones of the Province since the geographic distribution of these stands reflects distinct gradients of wet and dry sulphate, wet and dry nitrate and ozone deposition. Although this analysis does not preclude the influence of other non-climatic variables on the growth of sugar maple, it does provide circumstantial evidence of a relationship between air pollution and sugar maple growth.

Hardwood Nutrition Studies

A potential effect of acidic precipitation is the leaching of plant nutrients from the soil. This could result in reduced plant growth and be reflected in the foliar chemistry of vegetation growing on the site. A study was carried out in 1986 and 1987 to determine the foliage and soil chemistry for two tree species; yellow birch (*Betula alleghaniensis*) and sugar maple at 35 of the 110 Hardwood Decline Study sites for each species. The intent of the sampling was also to identify relationships between foliage and soil chemical properties and to relate these to the condition of the trees at the sites. The results of this study showed no marked nutrient imbalances or deficiencies in the foliage or soil samples. pH was the soil characteristic which was most consistently correlated with foliar element concentrations. Foliage of both species contained lower concentrations of calcium and magnesium and higher concentrations of manganese, copper and zinc when associated with lower soil pH. Higher Decline Index ratings were associated with lower soil pH and higher foliar manganese.

Early Diagnosis Project

A study comparable to one conducted in Europe in 1986 was initiated in Ontario in 1987. The intent of this study was to determine whether certain chemical tests might distinguish those trees which were suffering from stress prior to the development of dieback symptoms. Samples of sugar maple white pine (*Pinus strobus*) and Norway spruce (*Picea abies*) were collected across a pollution gradient from southwest to southeastern Ontario. Two tests (contact angle measurement and Hartel test)

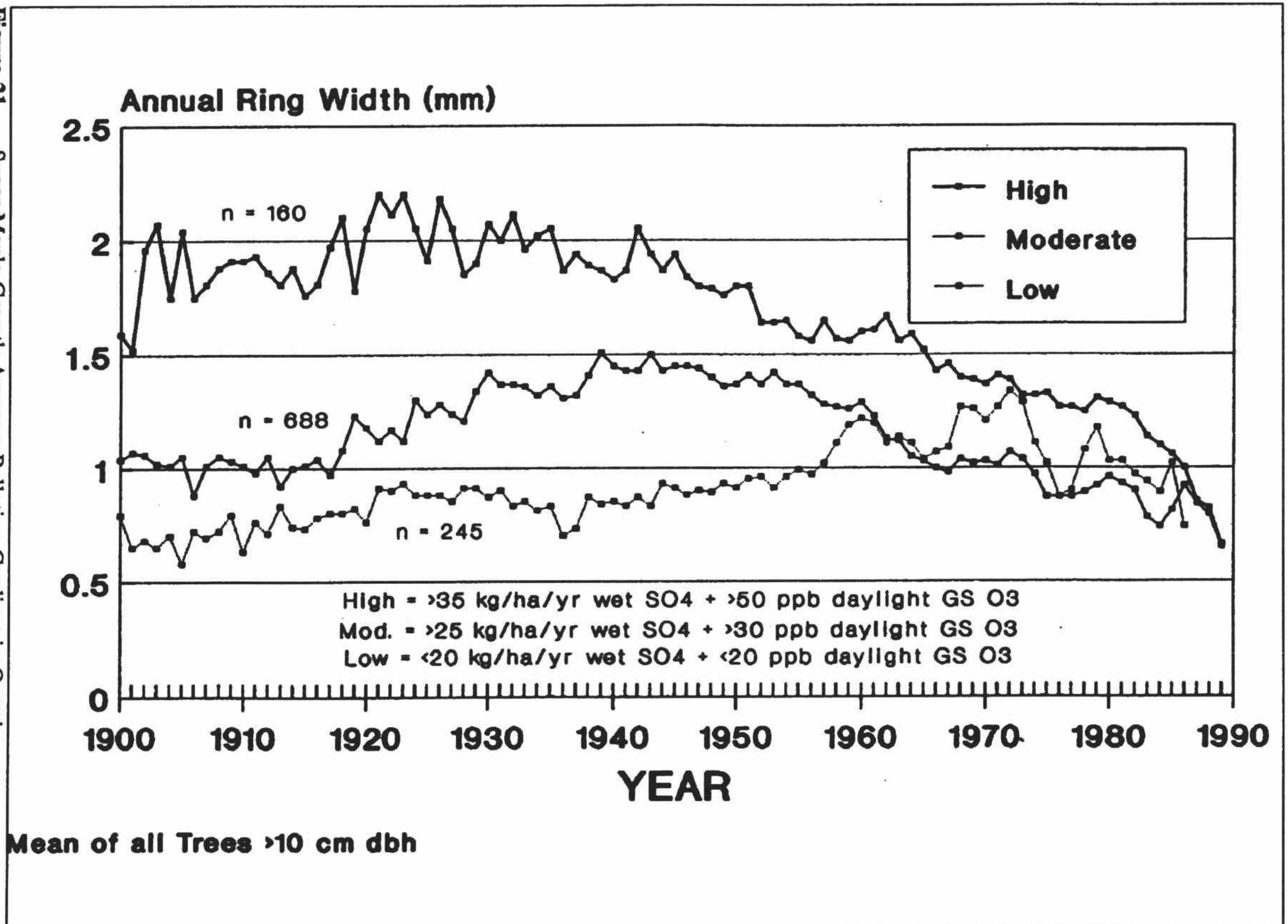

Figure 21 Sugar Maple Growth Across a Pollution Gradient in Ontario

performed on the conifer foliage suggest that both external and internal changes were occurring in the foliage. Foliage collected in the zone of highest pollution, measured as ozone or sulphate deposition, showed properties of older foliage earlier than did foliage collected in less polluted zones. Inorganic analysis of foliage tissues did not reveal marked deficiencies in nutrient concentration. Pigment content of maple and pine foliage collected in the field showed shifts with age of foliage (with pine) but the values were too variable to demonstrate statistically significant relationships with deposition zones.

Seedling trees of sugar maple, white spruce (*Picea glauca*) and 3 clones of hybrid poplar (*Populus euroamericana*) were subjected to simulated acidic precipitation (SAR) under the REC system. The pH's of the SAR were 3.2, 4.3 or 5.6. Samples of foliage were analyzed for pigments near the end of the growing season. No significant differences among six pigments or in their relative ratios were noted for maple or spruce. In the case of the poplar, the concentrations of all six pigments were lowest in the lowest pH treatment (pH 3.2). Various ratios of pigments were similar in all pH treatments. Differences among the poplar clones were greater than the differences caused by acidic treatment. In summary, the results of some chemical tests of tree foliage showed shifts in response to pollution stress; however, the inherent variability within each species precludes routine application of the tests utilized to date as a diagnostic test procedure. Under laboratory conditions, the tests may be more useful.

White Birch Dieback Study

A study of severe dieback of birch trees on the northeastern shore of Lake Superior and at other locations in northern Ontario is underway to determine whether anthropogenic stresses including acidic precipitation and fog were responsible for the observed dieback. Thusfar, studies have concentrated on sites south of Wawa. A complex of birch species (*Betula papyrifera* and *B. cordifolia*) is involved and there appears to be a shift in species composition over time. The surface soils are extremely acidic (to pH 2.9). Analysis of soil, foliage, roots and sap are ongoing.

Intergovernmental Cooperative Studies

A coordinated study of sugar maple decline across several provincial, state and federal jurisdictions in NE North America was set up in 1988. The purpose of the study was to monitor the rate of change of dieback of sugar maple in relation to different zones of pollution across the growing range of the species using a uniform assessment procedure. Over 100 permanent plots were established with 24 of these located within the Province of Ontario. The final field evaluations are scheduled for 1990 but the project may be continued as part of other programs.

Implications

The information obtained from the studies noted above indicate that the nature of the problem for the terrestrial environment is very complex. Involved are natural factors including climatic variables (especially drought), soil characteristics, insects and disease organisms. Forest stand management is important and acidic precipitation together with associated long range transported pollutants are considered to be additional stresses. It is highly probable that the latter are responsible for exacerbation of natural stresses to the detriment of the native vegetation.

The observations which are of the greatest concern are those related to the reduced

growth of the forest trees. If outwardly-healthy trees are reducing the volume of wood produced annually by approximately one half, this could have disastrous consequences for the production of quality hardwood and softwood lumber as well as maple syrup produced in these regions. Other forest uses in these regions that are jeopardized include recreation, tourism, wildlife habitat and aesthetics. Clearly, much greater efforts are required to determine whether these growth reductions are real and to assess the role of acidic deposition and associated long range transported pollutants as a causal or predisposing factor.

D. TERRESTRIAL WILDLIFE STUDIES

The wildlife studies conducted by the Ontario Ministry of Natural Resources were established to document the impact of acidic precipitation on selected wildlife species and long-term monitoring of selected biota. Most of the investigations to date have focused on cadmium movement in the food chain of several species of game animals.

Investigations continued in 1986/87 on cadmium levels in Ontario moose and deer in relation to forage and the soil's sensitivity to acidic precipitation. This study provided data on human health implications of consuming kidney, liver and muscle tissue of moose and white-tailed deer from Ontario. Cadmium levels increased with age in both moose and deer and varied regionally with the highest seen in the Algonquin Region. Cadmium levels for both species were elevated above World Health Organization standards and as a result of the study, the Ontario Ministry of Natural Resources in consultation with the Ontario Ministry of Health recommended that the public not consume kidneys or livers of Ontario moose or deer. A subsequent program to examine cadmium levels in Ontario black bear showed a similar pattern of cadmium concentrations to that seen in deer and moose.

Cadmium levels in three species of aquatic macrophytes preferred as forage by Ontario moose were investigated in 1988 and 1989 as one component of the program to determine mechanisms of cadmium uptake in the food chain. Two species (*Utricularia vulgaris* and *Potamogeton gramineus*) are highly palatable to moose while the third (*Nymphea odorata*) is not favoured. Cadmium levels in these species in Algonquin Park, a poorly buffered, acid sensitive area were significantly higher than those from well-buffered wetlands near Cornwall. Preferred forage species accumulated the highest cadmium levels.

In a cooperative study with Laurentian University in 1986-1988, cadmium levels were examined in the soils, forage and faecal pellets from white-tailed deer yards located in areas designated as either geochemically sensitive or tolerant to acidic precipitation. Cadmium levels were elevated within acid stressed soils of non-buffered sites. Direct positive correlations between plant and soil levels were observed for three species. Trembling aspen (*Populus tremuloides*) showed the highest cadmium concentrations. Cadmium levels within pellets were also elevated at non-buffered deer yards.

Biogeochemical Studies (Task 4)

Contact: P. Dillon

The purpose of the biogeochemical studies is to determine how catchments interact with acid deposition and how streams and lakes are affected as a result of these interactions. There are six subtasks being investigated which include Mineral Weathering, Soil, Bioaccumulation, Hydrological and Wetland Studies, and Watershed Manipulation Experiments.

A. MINERAL WEATHERING STUDIES

Primary mineral weathering is a source of secondary minerals, a source of essential nutrients for vegetation and is one of the major sources of acid neutralization in soils.

The following is a summary of the results of mineral weathering studies conducted in the Muskoka-Haliburton region over the last five years.

Weathering has created an aluminum-rich layer on the surface of feldspar grains in the B horizon of Plastic Lake soils. This layer may be responsible for some of the Al leached from these soils by acidic deposition.

The major minerals weathering in the Plastic Lake catchment are feldspars. Long-term weathering rates determined from depletion profiles of soil minerals are less than current denudation rates suggesting that current rates may be elevated by acidic deposition.

A study of the secondary mineralogy of Plastic Lake has been completed. Vermiculite dominates in the Ae horizon and the clay mineral suite in the B horizons is comprised primarily of hydroxy-interlayered vermiculite and kaolinite. Amorphous Al in the B horizon and aluminum present in the vermiculite interlayer, is likely the result of eluviation from upper horizons (Ae). These results agree well with and complement the soil/soil solution studies (see below).

Laboratory leaching studies on feldspar minerals using aqueous solutions, similar in composition to those found in the soil water of Plastic Lake, have demonstrated minimal leaching profiles on mineral grains similar to those found in the Ae horizons of Plastic Lake. This suggests that under field conditions, the feldspars are dissolving congruently.

Carbon isotopes (^{13}C and ^{14}C) were used as natural tracers to evaluate the sources and sinks of inorganic carbon in the Harp Lake catchment. Analysis of the C isotopes in dissolved inorganic C (DIC) showed that all of the alkalinity produced in the catchment and transported to the streams and/or lake (including the groundwater) resulted from silicate mineral weathering, not from any carbonate sources.

B. SOIL STUDIES

Soil/Soil Water Interactions

The chemistry of the leachate from 12 sets of lysimeter pits along with streams and

deep groundwater in the Plastic and Harp lake catchments was monitored over the last 5 years. The major difference in the stream chemistry between Plastic and Harp catchments can be accounted for by the long residence time of the groundwater at Harp Lake. The Plastic lake soils are very thin and there is comparatively little groundwater.

Differences in the unsaturated zone soil chemistry also occur and are important to vegetation and soil modelling of long-term changes in acidic deposition.

Harp soil solutions from the LFH (upper organic) horizons have greater alkalinity, pH and base cation concentrations than similar horizons at Plastic Lake. Leachate draining the soil from the B and/or C horizons have similar base cation concentrations. Furthermore, the concentration of inorganic SO_4^{2-} is much higher in the B horizons than the LFH/Ah horizons at both sites, but levels are higher in the Plastic Lake soils. These findings confirm that the differences in alkalinity and pH of water leaving the soils at the two sites are not due to differences in base cation release, but to differences in the release of aluminum and, more importantly, sulphate.

It is thought that the strong uptake of base cations in the B horizon at both sites is due to the large amounts of hydroxy-interlayered vermiculite present in these horizons. Higher levels of adsorbed SO_4^{2-} at the Plastic Lake site result in greater SO_4^{2-} release from the soils (see below). Aluminum release is also greater at Plastic Lake and is probably due to greater dissolution of vermiculite containing Al in the interlayer position, as well as allophane and other amorphous Al complexes.

Selective soil extraction studies at Dorset have shown that the major amorphous Al-Si compound in Plastic and Harp Lake mineral soils is 2:1 allophane. The Al component is in equilibrium with hydroxy- interlayered vermiculite. Soil solution concentrations of Al and Si agree well with this hypothesis. Future work will attempt to verify this hypothesis.

Sulphur Studies

Sulphur adsorption and release are the major geochemical reactions in soils that buffer acidic deposition. Greater levels of adsorbed SO_4^{2-} are found in Plastic Lake soils than in Harp Lake soils, and correspond well to the soil solution results reported above. Almost all the inorganic sulphur in the soil solution is SO_4^{2-} which has the same isotopic composition as organic sulphur. This suggests that it has passed at least once through the organic sulphur pool.

A new method for organic sulphur analysis has been developed by the Laboratory Services Branch, and will permit the accurate measurement of organic sulphur in small amounts of soil water from the lysimeters. This will allow quantitative estimates to be made of the relative importance of organic sulphur flux from the LFH to the B horizons. Its role in providing SO_4^{2-} through decomposition to the adsorbed SO_4^{2-} pool can then be identified.

Laboratory SO_4^{2-} adsorption and desorption studies have demonstrated the pH dependence of SO_4^{2-} adsorption/desorption processes. In the upper mineral horizons there is more adsorbed SO_4^{2-} and greater desorption under ambient pH conditions. This agrees well with the field results described above and will provide a quantitative basis for modelling this process.

C. BIOACCUMULATION STUDIES

The accumulation of base cations in vegetation can be a significant factor in a catchment's overall budget. This accumulation can also generate acidity in the soil. To evaluate these effects, trees over 10 cm in both the Harp 4-21 subcatchment and Plastic Lake PC-08 subcatchment have been cored for temporal dating and chemical analysis. The dating and chemical analyses have recently been completed and collected into a database awaiting interpretation.

D. HYDROLOGICAL STUDIES

Hydrology plays an important role in routing acidic deposition to various components of the landscape. These components have markedly different impacts on water chemistry. As previously mentioned, hydrological studies have demonstrated the importance of deep, long-residence time groundwater as a contributor to stream chemistry differences between the two basins. The relative importance of saturated overland flow (runoff) in different parts of the Plastic Lake catchment has also been documented. Water that has had little contact with the upland mineral soils or wetlands has a substantially lower pH and lower Al and Si concentrations. With the exception of the wetland areas during spring melt, most precipitation passes through the soil.

E. WETLAND STUDIES

Wetlands in a watershed have been reported to have a significant effect on nutrient budgets for the watershed by acting as nutrient sources and sinks and as the site of nutrient transformations. To investigate this, monthly and annual input-output budgets for total N, total organic N, total inorganic N, NH_4^+ nitrogen, nitrate, total P and DOC were measured on a total of five wetlands, located in Harp Lake, Plastic Lake and Paint Lake catchments.

The wetlands transformed N by retaining inorganic N and exporting an equivalent amount of organic N. Beaver pond wetlands retained NO_3^- and allowed NH_4^+ to pass through while conifer swamps retained both NO_3^- and NH_4^+. DOC fluxes into and out of the beaver ponds were equal but output from the conifer swamps exceeded input by greater than 90%. Phosphorus retention was less than 20% of the P input, and budget uncertainties were greater than or equal to retention rates.

Marked seasonal trends in nutrient retention were observed. Nutrient retention coincided with stream flow, increased evapotranspiration and biotic uptake during the summer. Net nutrient export occurred during the winter and spring when stream flows were highest and biotic uptake was low. Future work on the wetlands will concentrate on sulphur budgets and methane (CH_4) production.

F. WATERSHED MANIPULATION EXPERIMENTS

Catchment Manipulations

Environment Ontario has contributed to the funding of two catchment-scale manipulation experiments. The first and largest of these is the RAIN (Reversing Acidification in Norway) project. This experiment's purpose

is to examine the effects of changes in precipitation chemistry on soil and surface water acidification. At a pristine area in western Norway, two catchments are being acidified by the addition of H_2SO_4 and H_2SO_4 + HNO_3^- respectively. At an acidified catchment in southern Norway, ambient acid deposition is excluded by means of a roof and clean precipitation added underneath.

At the pristine sites, results over 5 years indicate that the acid-sensitive catchments responded rapidly to increases in acid deposition. Single severe episodes may be sufficient to acidify runoff to the extent that pH and Al levels are toxic to some fish. Chronic or long-term acidification of runoff resulted in only 3 or 4 years. Soil acidification also took place.

At the acidified sites, data from the acid-exclusion experiment showed that reductions in acid deposition result in decreased concentrations of strong acid anions in runoff. Soil acidification also began to reverse.

The changes in runoff and soil chemistry at the two study areas can be explained quantitatively in terms of the key soil-chemical processes including sulphate adsorption, cation exchange, weathering and organic acid buffering. The MAGIC model (discussed in the section "Aquatic Studies") successfully predicted the observed changes in runoff chemistry at both the pristine and acidified sites. The RAIN project shows that sensitive catchments respond to changes in acid deposition (increases or decreases) within a few years. The reversibility of the acidification process observed here agrees well with the studies of the Sudbury lakes and Plastic Lake (see Aquatic Studies).

Parts of the terrestrial uplands of Lake 302 in the Experimental Lakes Area near Kenora were also acidified. Nitrogen and sulphur deposition, within six small catchments and one larger catchment (8 ha in size), were manipulated to simulate southern Ontario deposition by spiking the snowpack with ammonium nitrate and sulphuric acid.

During the study, the hydrologic pathways and outflow generation pathways were monitored to explain the observed stream chemistry at different locations within the catchment. Two distinct landscape areas were monitored, bedrock areas with no infiltration of precipitation and forested areas with substrate infiltration in a soil depth averaging seven centimeters.

The bedrock sub-basin showed peak flows at snowmelt and during rain events. Aluminum levels peaked during these periods, and increased over time possibly as a result of different contributing areas during the runoff even. The forested sub-basin also peaks at these times but a lag effect was seen.

After acidification of one of the catchments, pH decreased by 0.2 units. Cations and sulphate also decreased initially before returning to control levels. In the control watershed, the inorganic Al fraction was one third to one half of the monomeric organic fraction. However, for the acidified watershed, until three weeks into the melt period, inorganic Al dominated.

Environmental Management and Economics Studies (Task 5)

Contact: J. Donnan

A. DAMAGES AND BENEFITS

Acid Deposition Effects models that were developed in the early 1980's were reviewed and, where possible, updated. These models are programmed in the Interactive Financial Planning System language which is supported on the Government mainframe computer network.

Five submodels were originally developed for the following receptor categories: Forests, inland lakes with commercial fisheries, agricultural crops, commercially harvested fur-bearing wildlife and materials and structures. Based on effects research to date, only three of the receptor categories are at some risk to acid deposition: forests, materials and structures and agricultural crops.

The 100 or so inland lakes with commercial fisheries in Ontario are generally very large with high alkalinity and buffering capacity and little risk of acidification. Research has revealed no verified dose-response relationships between acid deposition and fur-bearing mammals.

B. COSTS OF ABATEMENT AND MITIGATION

Sulphur dioxide and NO_x abatement technology and cost information for stationary sources in Ontario that was developed by consultants during fiscal 1988-1989 was applied to estimate the costs of achieving further emission reductions after the Countdown Acid Rain program is completed in 1994.

Costs were estimated to achieve reductions in total emissions of SO_2 and NO_x from stationary sources (including the four major corporate sources under regulation and the remaining industrial establishments which are not subject to specific regulations) by 10%, 30% and 50%.

Based on the type of technologies used for cost estimation at stationary sources only, further SO_2 reductions would be less costly per tonne removed than NO_x reductions.

Analyses of abatement cost functions indicate that the major regulated sources of SO_2 would be the most cost-effective sources for further emission reduction after the Countdown program has been completed. NO_x reductions at Ontario Hydro thermal power plants would also be the most cost effective for NO_x reductions at stationary

sources. Further reductions of NO_x by mobile and area sources (ie. the automobile and other internal combustion engines) were not considered in this study because reliable cost estimates of additional controls by automobiles were not available.

Another study had been proposed to investigate various approaches to reduce mobile source emissions beyond levels achievable with "on-board" emissions control devices. It was subsequently agreed to join with the Ministries of Energy and Transportation to carry out a more comprehensive effort.

A new terms of reference was completed and a competition initiated under the administration of the Ministry of Energy. The objective of the expanded study is to "identify and evaluate measures and approaches to reduce energy use, improve energy efficiency and reduce emissions in Ontario's transportation sector." A draft report is expected in February 1991.

Laboratory Support and Methodology Studies (Task 6)

Contact: F. Tomassini

Through the coordinated efforts of the Central and Dorset Laboratories, the Laboratory Services Branch (LSB) continued to support the various APIOS programs. A summary of the total test load for the program is shown in Figure 22. The 1989/90 fiscal year was a productive year and the highlights are:

A. INTERLABORATORY COMPARISON ANALYSIS

a. Eulerian Model Evaluation Field Study (EMEFS)

1. Precipitation - The LSB received 10 submissions (100 samples) for major ions, nutrients, physical parameters analyses in addition to a single submission for low level anions (chloride, nitrate and sulphate analyses). (Workload 2251 tests).

2. Air Filters - In January 1990, a new study was initiated on air sampling filters of nylon, Teflon and Whatman 41. The first submission of 72 filters was analyzed for ammonium, nitrate, sulphur dioxide

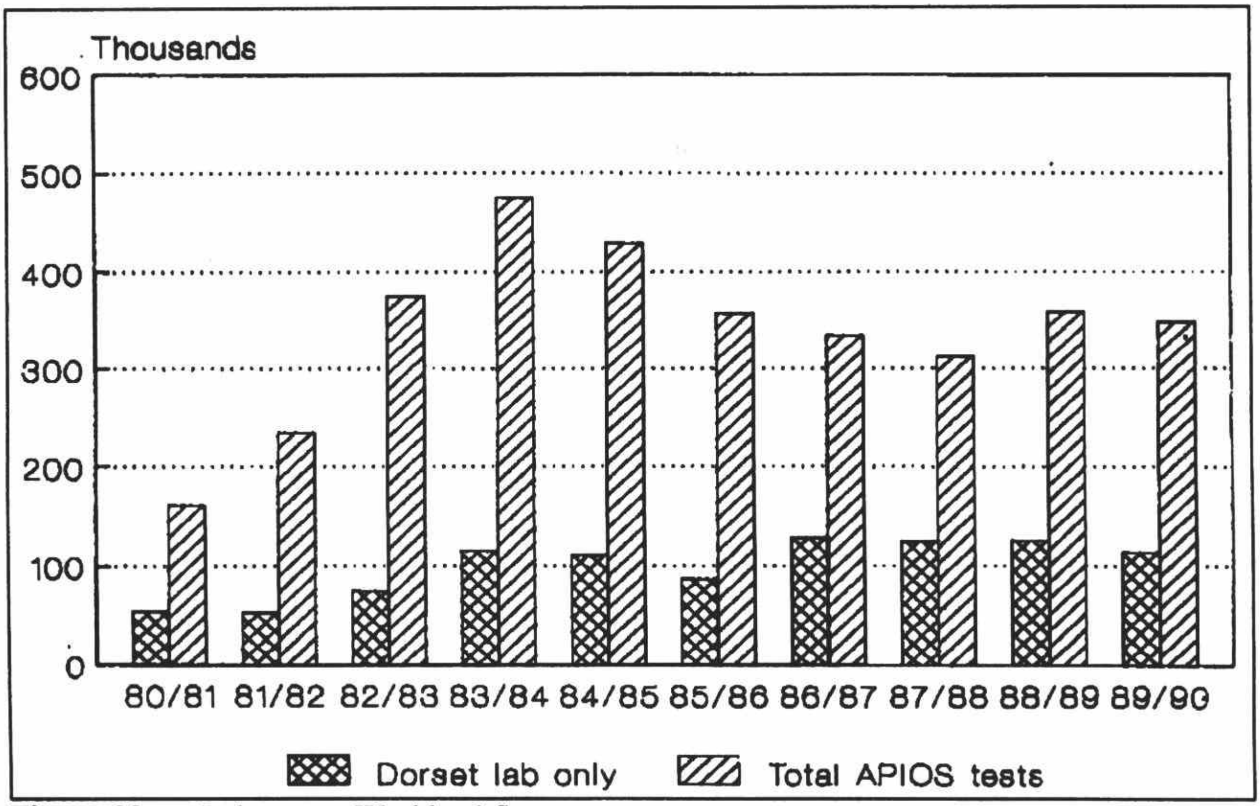

Figure 22 Laboratory Workload Summary

and sulphate. (Workload 126 tests).

b. Long Range Transport of Atmospheric Pollutants (LRTAP)

The Branch received 4 submissions (40 samples). (Workload 800 tests).

B. ANALYTICAL METHODOLOGIES

- Developed an ion chromatographic (IC) method for ammonium and conducted comparative analysis between IC and colourimetry.
- Compared the manual Klett-Summerson method for colour with the automated method.
- Investigated the effect of removal of sodium azide in the dissolved oxygen analysis.
- Established working group with the Water Resources Branch (WRB) to develop a large batch method for low level Cd, Cu, and Zn in water samples.
- Conducted pilot study for the preservation of water samples with sulphuric acid for total phosphorus, nitrate plus nitrite, ammonia-nitrogen and total Kjeldhal nitrogen analyses. The results, obtained from May to October 1989, were used to design a sampling procedure for precipitation samples in remote areas of Canada.
- The WRB in Dorset did a study on extractable Al, Fe and Si to gain an understanding of the secondary weathering products in the Plastic Lake soils.
- Refined the Bray II extractable method for P and used it for the analysis of available P for plants in the soil fertilization study conducted by the Air Resources Branch (ARB).

Abatement

Contact: A. Deshpande

Since 1980, Ontario has been convinced that sufficient evidence existed to implement immediate SO_2 controls while research continued to evaluate the benefits of these controls. As a result, Ontario was the first jurisdiction in North America to mandate emission controls based solely on the effects of long-range transport of air pollutants, as distinct from local ambient air quality standards. Ontario Hydro and Inco were the targets of these controls.

In the absence of an acid rain agreement with the United States, the federal government and the seven Canadian provinces east of Saskatchewan decided in March 1984, to take unilateral action and reduce their sulphur dioxide emissions to a ceiling of 2.3 million tonnes by 1994, a 50% reduction from the 1980 base case value. On February 5, 1985, they agreed to the series of steps to achieve the first 1.9 million tonnes of this reduction and committed to determining the allocation of any further reductions in sufficient time to achieve the 1994 objective.

Subsequently, Ontario went beyond the commitment it made at the federal/provincial meetings with its announcement of the Countdown Acid Rain program in December, 1985. Ontario will reduce sulphur dioxide emissions from the 1980 base case level of 2,194 kilotonnes to 885 kilotonnes by 1994.

Four non-appealable regulations were issued to the four corporate sources which together emit over 80% of Ontario's SO_2: Inco, Falconbridge, Algoma Steel (Wawa) and Ontario Hydro. The regulated annual emission limits were developed in consultation with each of the major pollution sources to ensure that economic activity need not be excessively inhibited.

A phase-in approach is taken. Regulated limits are summarized below (Table 4).

Table 4

Legal SO_2 Limits for the Four Countdown Companies

Source	Legal Limits Sulphur Dioxide (kilotonnes/year) 1986	1994
Inco, Sudbury	685	265
Falconbridge, Sudbury	154	100
Algoma, Wawa	180	125
Ontario Hydro, province-wide	370	175

A fifth regulation limits SO_2 emissions from new or modified boilers by placing a 1 per cent sulphur content constraint on the fuel or by requiring that an equivalent amount of SO_2 be removed from the flue gas.

In addition to the SO_2 limit, the sum of SO_2 and NO emissions is also regulated for Ontario Hydro.

Furthermore, there are interim limits by 1990 (Table 5).

Table 5
Ontario Hydro's Acid Gas Limits

	SO_2 (kt/yr)	SO_2 + NO
1986-1989	370	430
1990-1993	240	280
1994 +	175	215

Control technologies were not specified. The individual companies may choose their method of abatement. Regulations only require that the legal limits are met by the specified dates. The regulations provide for a three year research and development period. Semi-annual interim progress reports are required from the four major companies, for review by experts from various Ontario ministries. A final report of this Research and Development phase was received on December 31, 1988 from each of the metallurgical companies and in January, 1989 from Ontario Hydro. These reports clearly stated that the companies would be able to meet the regulated limits, and the reports described precisely how the emission limits would be met. The government's summary and analysis of all these reports along with copies of the companies' reports are available upon request. Semi-annual progress reports of the implementation phase continue to be required of the companies.

All four regulated sources met sulphur dioxide and acid gas (SO_2 + NO) emission limits in 1989 and Falconbridge Nickel Mines and Algoma Steel (Wawa) sulphur dioxide emissions were even below the 1994 targets (Table 6).

Table 6
Estimated 1989 Emissions

Source	1989 Emissions (kilotonnes/yr) SO_2	SO_2 + NO
Inco, Sudbury	637	-
Falconbridge, Sudbury	68	-
Algoma, Wawa	71	-
Ontario Hydro, province wide	305	368

An emissions verification program was developed for the four regulated sources by the Countdown Emissions Verification Subcommittee in 1989 and appropriate orders were issued to the companies in June-July, 1990. These orders require the companies to upgrade their sampling and analytical techniques to American Society for Testing and Materials (ASTM) or equivalent standards and to develop manuals which will form the basis of future emissions reporting systems. Companies will also look into implementing continuous emission monitoring techniques for SO_2 and NO if found suitable. Ontario Hydro will use an acceptable Continuous Emission Monitoring (CEM) system after completing a study comparing combustion gas monitors and in-stack monitors before 1992.

In the meantime all four companies will continue to use mass balance methods to estimate emissions and reported SO_2 and NO emissions will be subject to procedural audit by an independent consultant. These audits are expected to commence in January 1991 and will cover part or all of 1990 reported SO_2 and NO emissions.

Legal Initiatives

Contact: B. Carr

Under the Memorandum of Intent signed in 1980, Canada and the United States agreed to enforce existing laws and regulations responsive to the problems of transboundary air pollution. Since 1981, however, the Unites States Environmental Protection Agency has proposed the approval of revisions in State Implementation Plans (S.I.P.'s) under section 110 of the U.S. Clean Air Act which would lead to increases in allowable sulphur dioxide emissions from coal-fired power plants. Because increased emissions could affect the province's environmental quality, Ontario encouraged the U.S. EPA Administrator and state governments to disapprove any S.I.P. revisions which would result in any increase in permissible emissions of SO_2 in the U.S.

Seeking an avenue to effective acid rain controls in the U.S., Ontario has appeared at numerous hearings and participated in judicial proceedings since 1981. These efforts have been made in conjunction with several American states, environmental interest groups, and citizens, and are directed at two key parts of the Unites States Clean Air Act: sections 115 and 126.

Section 126 provides for the intervention of the EPA's Administrator in instances when interstate air pollution can be shown to be preventing the attainment of national ambient air quality standards, or interfering with those prevention of significant deterioration or visibility measures which the Act requires to be included as a part of a state implementation plan. This path to acid rain controls was blocked in March 1989, when the U.S. Supreme Court said it would not hear an appeal by a group of northeastern states. This decision means that an earlier ruling by the U.S. Circuit Court of Appeals will stand. That court ruled the EPA was not required to force reductions of pollution originating in other states since the northeastern states did not adequately support their claims of injury.

Ontario has been more actively involved in proceedings dealing with the internationally-oriented section 115. This section provides, in pertinent part:

(a) Whenever the (EPA) Administrator, upon receipt of reports, surveys or studies from any duly constituted international Agency has reason to believe that any air pollutant or pollutants emitted in the United States cause or contribute to air pollution which may reasonably be anticipated to endanger public health or welfare in a foreign country ... the Administrator shall give formal notification thereof to the Governor of the state in which such emissions originate.

Under section 115 the Administrator also has to make findings that the foreign country offers reciprocal rights to the United States and that the environment of the foreign country is endangered.

In April 1988, Ontario and a group of states filed independent petitions to the U.S. EPA Administrator requesting that he reaffirm findings of endangerment and reciprocity under section 115 which had been made informally in 1981 by the then Administrator

of the EPA and accepted in previous court actions, by publishing the findings in the Federal Register and then promulgating the findings as final rules.

In October, 1988 the Acting Assistant Administrator of EPA's Air and Radiation Office issued a response stating that it would be premature for EPA to take any rule making action until National Acidic Precipitation Assessment Program (NAPAP) findings are available for assessment in 1990.

In November 1988, a follow-up legal petition was submitted to the U.S. Circuit Court of Appeals for the District of Columbia by Ontario to require the U.S. EPA to formally make the findings of reciprocity and endangerment. The suit was joined by two U.S. environmental groups, the Sierra Club Legal Defence Fund and the Izaak Walton League of America.

Later, the Federal Government, Quebec, New Brunswick, and other Canadian environmental groups requested to become Amici Curiae (friends of the court) in support of Ontario.

Also in November, 1988, nine states (6 New England states, plus New York, New Jersey and Minnesota) also filed a parallel lawsuit against the U.S. EPA to force the agency to take similar action under section 115. This was joined by the National Audubon Society. Once formal findings are made the EPA is then required to compel states from which offending emissions come to reduce their emissions.

Arguments were presented to the Appeals Court in early 1990 and a decision has yet to be rendered.

Communications Initiatives

Contact: G. Merchant

The Communications Work Group (with representatives from the Ministries of Intergovernmental Affairs, Tourism and Recreation, and Natural Resources) discussed and implemented strategies to increase awareness of acid rain among Ontario and American residents.

ONTARIO

Ontario citizens are aware of acid rain as evidenced by the number of information requests made directly to the APIOS Coordination Office. Some 590 individualized packages of information were mailed in this fiscal year and about 400 people visited the Office. As well, there were numerous presentations to schools and interest groups. These contacts are over and above the enormous quantity of requests for acid rain information handled directly by the Communications Branch.

UNITED STATES

The communications strategy directed toward targeted U.S. audiences stresses several themes:

- Human Health - Evidence has been accumulating that acid gas emissions are detrimental to human health.

- Effective American Legislation Is Required - There is a need for an interim and longer-term cap on acid gas emissions to prevent back-sliding and reach a scientifically supportable critical loading.

  The sooner the better as biological systems tolerate considerable stress before suddenly crashing.

- It Can Be Done - Cost-effective acid rain abatement technology options do exist. There are specific economic benefits of abatement in the creation of a strong environmental protection industry.

  Ontario's <u>Countdown Acid Rain</u> program serves as a model of a successful abatement program.

This approach is intended to increase Americans' awareness of acid rain and of the solutions that do exist. To this end, several initiatives were undertaken and included the following:

Ontario was the lead agency in organizing 2 acid rain information trips to Canada for a total of 13 congressional legislative assistants. These included visits to Toronto, the Dorset Research Centre, Sudbury, Ottawa and Quebec. Meetings were held with federal and provincial political leaders, the Canadian Coalition on Acid Rain, and executives from Inco, Ontario Hydro and Falconbridge.

Similar to previous years, an updated acid rain display was exhibited and staffed by

Ministry personnel at certain international conferences and shows. In a joint efort, the display was also loaned to various Canadian Consulates in the U.S. so that its impact could be felt in a greater number of venues. The display was well-received at all shows and many valuable contacts were made.

Endeavours to cultivate relationships with grassroots organizations continued as Ontario provided such groups with acid rain information to be distributed to their members.

APPENDIX I

INTERNATIONAL LRTAP PROJECTS - MOE CO-FUNDING

Project Title	Funding Agencies	Purpose
Acid Deposition and Oxidants Model (ADOM)	Environment Ontario Atmospheric Environment Service Umweltbundesamt (West Germany) Environment Quebec State of Minnesota State of New York Electric Power Research Institute	To improve predictions of source/receptor relationships, through the use of eulerian concepts.
Multi-bilateral Hardwood Decline Study	Environment Ontario Ministry of Natural Resources Forestry Canada Department of Natural Resources - New Brunswick Ministry of Energy and Resources - Quebec Northeast Forest Cooperative	A study of the etiology of sugar maple decline across northeastern North America using a common methodology.
Eulerian Model Field Verification	Environment Ontario Environment Canada U.S. E.P.A. Electric Power Research Institute	A study design has been prepared for the field verification of the eulerian model.

Project Title	Funding Agencies	Purpose
Reversing Acidification in Norway - NIVA	Environment Canada Norway Sweden Environment Canada United Kingdom	To test hypotheses on watershed sensitivity and to measure watershed response to reductions and increases in acid loadings. This issue has been recently raised by the U.S. E.P.A. as an impediment to designing a control program.
Unified Acid Deposition Data Base for Eastern North America	Environment Ontario Environment Canada National Atmospheric Deposition Program, U.S. Geological Survey Battelle Pacific Northwest Laboratory, U.S.	The unified data base should be useful for mathematical model evaluation and historical trend analysis.

INTERNATIONAL LRTAP PROJECTS - MOE PARTICIPATION

Project Title	Participating Agencies	Purpose
BACG (Bilateral Advisory Consultative Group)	Environment Ontario Ministry of Energy Ministry of External Affairs Environment Quebec Environment Canada Office of the President Department of State, U.S. Department of Energy, U.S. U.S. EPA National Acidic Precipitation Assessment Program	To respond to or implement recommendations of the Special Envoys' Report
Fisheries Loss Assessment Program	NAPAP Environment Ontario Ontario Ministry of Natural Resources EPRI Environment Canada	To assist NAPAP in the design of a program to assess fisheries loss in the U.S. related to acidic deposition.
Informal Calibrated Watershed Modelling Group	Environment Ontario Environment Canada United States Norway Sweden	To compare results and ideas on watershed studies. The work defines effects of acid rain and develops target loadings to prevent damage.

Project Title	Participating Agencies	Purpose
Interlaboratory Quality Assurance	Government and private laboratories in Canada and the U.S. (over 50 labs involved, including MOE)	To ensure the validity and compatibility of all data collected under LRT programs in North America.
Lake Acidification Mitigation Program	EPRI Clarkson College Environment Ontario	MOE has been requested to provide advice and information concerning lake liming projects.
National Acidic Precipitation Assessment Program Review	Environment Ontario Environment Canada Fisheries and Oceans Canadian Forestry Service Government and private laboratories in U.S.	Review of NAPAP interim assessment document findings.
National Surface Water Survey	EPA Environment Ontario Environment Canada	To characterize current water chemistry of lakes and streams in five U.S. Regions. MOE has been requested to assist in the development of the survey design.
Ontario/Germany Memorandum of Understanding	Ontario Federal Republic of Germany	To exchange information, scientists and modelling results to ensure that similar methodologies are used so that final results may be compared.

Project Title	Participating Agencies	Purpose
Ontario/Michigan Memorandum of Understanding	Ontario Michigan	To exchange information and perform joint studies to achieve and maintain a quality of environment to protect human health and the ecosystem where activities of one jurisdiction may affect the environment of the other.
Ontario/Minnesota Memorandum of Understanding	Ontario Minnesota	To exchange information on acid rain; to cooperate on specific projects (atmospheric modelling, RAIN - NIVA, aquatic effects in a medium deposition area).
Ontario/New York Memorandum of Understanding	Ontario New York	To exchange information on acid rain to improve understanding of acidification of the environment and establish coordinated courses of action in order to encourage abatement measures on an international basis.
Ontario/China Memorandum of Understanding	Ontario China	To exchange acid rain/LRT information, to develop and collaborate on acid rain/LRT projects, and to provide training for scientists through projects participation.

Project Title	Participating Agencies	Purpose
Ontario/NADP Intercomparison Study - Ely, Minnesota	Ontario National Atmospheric Deposition Program	To improve comparability of data.

APPENDIX II

APIOS RELATED TECHNICAL REPORTS, DATA REPORTS, AND SUBMISSIONS

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